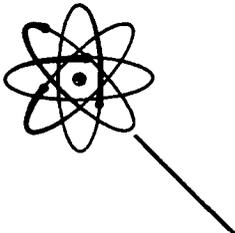


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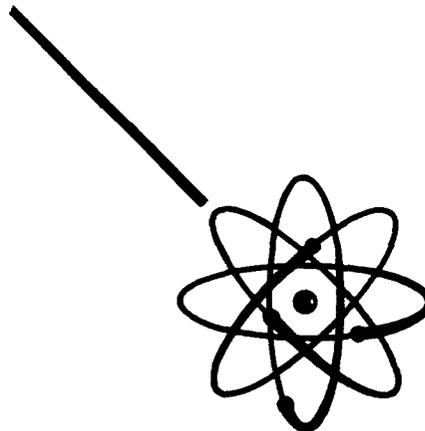
# **THE INDUSTRIAL ATOM**

**THE FIRE PROPERTIES OF METALLIC URANIUM**

**A Monograph By**

**R. B. Smith**

**April 12, 1956**



**Technical Information Service  
Atomic Energy Commission  
Washington, D. C.**

**ABOUT THIS MONOGRAPH:** Under suitable conditions, uranium is capable of self-sustaining combustion in air,  $N_2$ ,  $O_2$ , and  $CO_2$ ; the reaction is accompanied by emission of very intense but localized heat and light. Factors contributing to uranium pyrophoricity are discussed and a reaction mechanism involving surface hydride formation is outlined. Uranium-water reactions were also investigated.

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III

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TID-8011

## THE FIRE PROPERTIES OF METALLIC URANIUM

by R. B. Smith

In general, the fire properties of metallic uranium are quite similar to, but more pronounced than, those of magnesium of similar form. Under suitable conditions, uranium is capable of self-sustaining combustion in air, nitrogen, oxygen, and carbon dioxide. Investigators are agreed that the reaction at high temperatures between steam and uranium is much more vigorous than that between uranium and oxygen. While quite rare, severe spontaneous fires have been experienced during handling of fine moist powders under water as well as under inert gas (helium and argon). Burning uranium is capable of violently reacting with all commonly used fire extinguishants.

Self-sustaining combustion of uranium in air is accompanied by emission of very intense but localized heat and light. The rate of combustion may range from explosively violent, when finely divided metal powders are dispersed in a dust cloud, to very slow quiescent burning in the case of massive metal. Complete self-sustaining combustion of a one-inch diameter rod, for example, may require up to 24 hours before all of the metal is ultimately converted into the oxide. The metal burns without flame. The heat created during a fire has often been sufficient to burn holes through supporting thin sections of steel or cast iron. Once initiated, the rate of metal combustion is little affected by the porous oxides that are created.

That the pyrophoricity of metallic uranium can vary over an extremely wide range is attested to by the many vagaries experienced to date. For example, it is often possible to heat massive pieces of metallic uranium in air to temperatures up to the melting point without initiating oxidation at a rate sufficient to obtain self-sustaining combustion when the heat source is removed. By contrast, one case was reported in which a "yeast cake" size piece of uranium ignited spontaneously while resting on a block of dry ice!

Before further discussion of the vagaries in uranium pyrophoricity, it is important to point out that:

1. Massive uranium is normally incapable of self-sustaining combustion in air without the continued application of heat from external

sources. The occurrence of fires during rolling, extrusion, forging, storage, etc., is exceedingly rare even when manufacturing sheet uranium strip.

2. The frequency of fires that may be expected while metallic uranium is being sawed or while lathe or milling operations are being performed will vary with the type and rate of coolant application.

a. If no coolant is used, the chips or sawdust will often spontaneously ignite in air at the time of their creation.

b. If mineral oil coolants are liberally applied during machining operations, few if any spontaneous fires may be expected to occur in the chips.

c. If water (or a water soluble vegetable oil) is copiously applied as coolant, sporadic minor fires may be expected in the metal chips or sawdust at the time of their creation. Such a fire in a single lathe chip normally "burns itself out" after a few seconds duration but may on occasion spread to other chips in the immediate area. When this occurs in a small amount of lathe turnings, the fire may generally be easily extinguished by "dousing with coolant." A fire involving a large amount of chips on a lathe bed may be difficult or impossible to extinguish. While uranium chips, turnings, and sawdust do not normally spontaneously ignite during subsequent handling in air, literally thousands of spontaneous fires of this type have been suffered. Extinguishment or control of such fires is difficult or impossible where large accumulations of chips or turnings are involved. Controlled incineration or digestion by chemical means afford the safest methods for averting fire hazards from uranium chips and turnings. Where this is not possible, care should be taken to keep accumulations of metal chips at any one point to a minimum. The hazards of spontaneous ignition may be minimized by storage of chips under mineral oil although under these conditions gaseous hydrogen will be very slowly evolved. When chips are stored under water (or water soluble coolants), hydrogen evolution takes place somewhat more rapidly.

3. Finely divided metal powders created by any means are particularly pyrophoric. Grinder sludge, for example may spontaneously ignite while immersed in water for periods of a day or more. Dry uranium powder normally ignites spontaneously when exposed to air. If the metal powder is dispersed in air, spontaneous explosions may be expected. A detailed investigation into the hazards of uranium powder has been reported by Hartman (Bureau of Mines Report No. 3202).

#### VAGARIES IN URANIUM PYROPHORICITY

Unusually high pyrophoricity may be displayed in uranium either as a purely surface phenomenon or less frequently as a uniform character-

istic of all metal in the specimen. The following incidents will serve to illustrate some of the vagaries experienced to date:

1. Incidental to the manufacture of metallic uranium powder, some of the powder was slowly accumulated in a sump under perhaps 20 feet of sea water. Such accumulations are understood to have built up for periods of a month or so without incident following which, without prior warning, a vigorous reaction took place resulting in creation of a local "geyser." Following such eruptions, no further eventful happenings occurred until subsequent accumulations of metal powder permitted repetition of the preceding event.

2. Specimens of massive, low density (e.g., density of 15) metallic uranium are reported to have spontaneously ignited in air after having rested without incident for several days on a metal shelf.

3. The external jackets on several experimental uranium fuel elements failed after exposure in a water-cooled reactor. When the jackets of these fuel elements were removed by remote control means (they were highly radioactive), existence of powdered material not present when the elements were fabricated was noted. The powder spontaneously ignited when exposed to air.

4. Following the hot rolling of uranium billets into rods, the rods (roughly 1 inch in diameter) were placed on a straightening rack where they were permitted to cool to room temperature. Occasionally a rod would be encountered which not only failed to cool but instead would slowly increase in temperature until self-sustaining combustion was initiated and continued for approximately 24 hours until all of the metal had burned to the oxide. While numerous and simultaneous changes made during this period preclude pinpointing reasons for the subsequent significant decrease in the number of "pyrophoric" rods encountered, the presence of metal contaminants (particularly iron and copper) was suspected to be a major causative factor.

5. A large number of massive uranium pieces approximately one-half inch thick was unintentionally exposed to heat from an external fire of approximately one hour duration. The uranium ignited and continued to burn (without the application of heat from any external source) for almost a day. It should be pointed out that it is normally impossible to obtain self-sustaining combustion of massive uranium metal without continued application of external heat.

6. Prior to anticipated shipment, scrap uranium metal was incinerated to convert it into the oxide following which it was cooled to room temperature prior to packaging. When the "oxide" was received several days later at its destination, fire broke out in the packing material. Investigation revealed pieces of burning metal within the oxide.

7. Moist powdered uranium on steel trays was placed on cast iron, steam heated shelves of a vacuum dryer. After purging with an inert gas (argon) and application of vacuum, a spontaneous fire would occasionally occur of such intensity as to burn holes through both the supporting tray and the supporting cast iron grid.

8. A series of spontaneous fires had occurred in compacted uranium chips stored in closed-top 55-gallon metal drums under outside weather conditions. An investigator opened one such drum and, after noting that the drum contents were at room temperature and that everything appeared to be normal inside, was warned by an operator to "stand back." About 15 seconds later, the investigator witnessed a column of fire shoot up roughly 25 feet into the air and then immediately subside. Following this, it was found that the entire contents of the drum were incandescent.

9. Loose, dry uranium chips had been stored for about 6 months at room temperature in small (about 2 quarts) stainless steel containers having loose-fitting lids. On Saturday, a guard observed that in the area of these cans the temperature had risen to 105°F and called a supervisor who inspected each of the cans, noting nothing unusual, and left after correcting conditions which had led to previous overheating of the room. Temperature in this room over the weekend was at normal levels. Monday morning the supervisor again inspected the cans and this time found that many of them showed evidence of having been exposed to a high temperature. In part, this evidence consisted of holes that had been burned in the sides of the cans. Investigation revealed that while fire had unquestionably occurred in the uranium within the cans, only 15 percent of the metal (on an average) had been converted into the oxide.

10. During experimental research, it was found that two small batches of uranium powder appeared to have been rendered "nonpyrophoric" after treating the powder with carbon tetrachloride. When this was tried on a third and larger batch, a violent explosion occurred severely injuring the person conducting the experiments.

11. After removal of loose uranium chips from a nitric acid bath, small localized spontaneous fires have been occasionally encountered. When such fires occur in the open, they have on many occasions been extinguished by applying water with a hose. When fires of this type occur under confined conditions, they have been extinguished by flooding with CO<sub>2</sub> or in extreme cases with argon (although in rare cases even the latter fails to provide extinguishment of the blaze). On several occasions, localized fires in chips wet with nitric acid (handled in the open) have not only spread throughout the chips in the container, but have also been accompanied by explosions of sufficient violence as to seriously damage adjacent walls and ductwork.

12. Uranium powders (which are normally prepared by pyrolysis of uranium hydride) are usually highly pyrophoric. That the pyrophoricity of such uranium powder may vary over quite a wide range is fairly well recognized. (In this connection, it is of interest to note that catalytic iron prepared by hydrogen reduction of iron formate at elevated temperatures spontaneously ignites when suddenly exposed to air at room temperature.) Various patents have been issued for "stabiliza-

tion" of pyrophoric metal powders such as patents 2,677,668 and 2,677,669 issued to J. E. Ahlberg in May 1954.

13. It has previously been pointed out that spontaneous fires have been reported to have taken place while uranium powder or chips were submerged in water. Conversely, however, many fires involving incandescent uranium chips have been extinguished by immersion of the chips into water. Similarly, it is known that self-sustaining combustion of uranium can take place at elevated temperatures with  $\text{CO}_2$  but despite this fact operators have occasionally been successful in extinguishing uranium fires using  $\text{CO}_2$  extinguishers.

While the above list does not exhaust the types of vagaries experienced to date during handling of metallic uranium, it is perhaps sufficient to partially illustrate the basis for the following tentative conclusions which were reached after study of the available circumstantial evidence involved in hundreds of uranium fires:

1. Unusually high uranium pyrophoricity may be acquired as a consequence of factors associated with the initial preparation of the metal (this point has been confirmed by research in the case of metallic plutonium) or it may be acquired at a later period as a consequence of metal exposure to air and moisture under quiescent conditions.
2. The presence of certain metal contaminants or existence of the metal in a stressed condition (e.g., lathe chips) tends to increase the probability that a uranium specimen will acquire added pyrophoricity after initial formation of the metal.
3. Spontaneous ignition of uranium involves a rapid initial exothermic surface reaction with air or water. As contrasted to the better known cases of spontaneous ignition (e.g., oily rags), the spontaneous ignition of uranium does not appear to be preceded by any slow, gradual build-up of temperature.
4. Uranium pyrophoricity is strongly influenced by the nature of the surface oxide. Surface oxides acquired during metal exposure to dry air appear capable of very materially reducing (if not eliminating) the ability of a metal specimen to acquire added pyrophoricity. If other factors are equal, the degree of pyrophoricity increases with increasing surface to mass ratio of metal specimen.
5. In common with many other metals, uranium combustion in air is promoted by the presence of moisture in limited amounts.
6. Low density (e.g., apparent density of 15) specimens of uranium metal prepared by powder metallurgical means through decomposition of compressed uranium hydride are believed to have been involved in most if not all known fires in which massive pieces of metal spontaneously ignited while at room temperatures. It should be noted, however, that metal produced in vacuum casting operations or metal prepared by bomb reduction means will also occasionally display unusually high pyrophoricity particularly at the metal surface.

## THEORIES AS TO THE MECHANICS CONTRIBUTING TO URANIUM PYROPHORICITY

Any explanation as to the mechanics causing unusually high pyrophoricity in metallic uranium must take into consideration the conclusions detailed in the preceding section. Study of uranium fires experienced to date affords particularly strong circumstantial evidence on the following points:

1. Metal exposed to moisture under quiescent conditions (and preferably in the presence of air) appears to be almost invariably necessary before uranium can acquire unusually high pyrophoricity.
2. The probability that a particular uranium specimen will be unusually pyrophoric is greatly increased when the metal exists in a stressed condition and/or when it contains certain metal contaminants.
3. Unusually high pyrophoricity may exist either as a purely surface phenomenon or, less frequently, may be a property characteristic of all metal in the specimen.

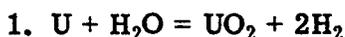
The ability of moisture to affect metal pyrophoricity was dramatically displayed in the following incidents:

1. An AEC contractor who handled uranium chips in dried air very rarely encountered any incidence of spontaneous fires. When the air dryer failed, a relatively large number of spontaneous fires was experienced. When the air dryer was repaired and replaced in service, the incidence of such fires again fell to practically zero.
2. The following incident provides unique and spectacular evidence that pyrophoricity of metals other than uranium may also be markedly influenced by exposure to moisture. Trouble was experienced in getting a reaction of gaseous zirconium tetrachloride with metallic magnesium to go to completion. When the furnace was opened, a thin surface crust was found which was thought to consist of a mixture of zirconium, magnesium, and magnesium chloride. A piece of this crust, roughly one square foot in area removed for inspection and test, appeared to be inert (i.e., no sparks) when scratched with a file and hit with a hammer. The piece was next placed under water for the purpose of leaching out the  $MgCl_2$ . This operation was repeated three or four times without any unusual occurrence. During the last attempted extraction and without prior evidence of any reaction, a violent explosion occurred (while the piece was under water) which shattered the laboratory bench, blew out a window some 20 feet away, and seriously injured the investigator. During subsequent cleanup, particles of metal found in adjacent plaster walls spontaneously ignited when the surface layers were touched with a brush. This incident is considered significant in strengthening the circumstantial evidence that:
  - a. Under suitable exposure conditions, metal pyrophoricity can be materially increased by exposure to water.

b. Spontaneous metal fires need not be preceded by any slow buildup of heat.

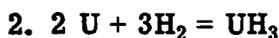
c. A connection probably exists between factors causing added metal pyrophoricity and factors involved in water-metal explosions.

If one accepts the premise that uranium pyrophoricity can be increased as a consequence of metal exposure to water under suitable conditions, the following known reactions must be considered:

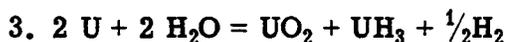


There is no evidence that this exothermic reaction has ever taken place at room temperature at a rate sufficiently rapid to cause significant heating of the metal. It is known that at room temperature hydrogen-saturated water corrodes uranium much more rapidly than water saturated with oxygen. Both conditions may simultaneously exist when the metal is exposed to quiescent moist air, thus introducing electrolytic factors, the net affect of which has not been extensively studied. Only one of the many investigators of the reaction reported (in 1943) that the  $UO_2$  produced was pyrophoric (see pp. 303 and 309, Vol. VIII-5, of the National Nuclear Energy Series).

While additional heat from hydrogen combustion may increase the rate at which uranium burns in air, this cannot account for the uranium fires that have taken place in the absence of air. Thus, there is little, if any, evidence that the subject reaction in itself can promote unusually high metal pyrophoricity.



This reaction has also been extensively investigated and could conceivably occur as a consequence of hydrogen evolved in reaction 1, above. The  $UH_3$  is brittle, extremely finely divided and highly reactive with both air and water (although this reactivity is quite erratic where small amounts of  $UH_3$  are involved). There is little reason to believe this reaction could occur to any significant degree except under quiescent conditions tending to prevent immediate conversion of any  $UH_3$  formed to  $UO_2$ . Once initiated, the uranium-hydrogen reaction is quite rapid even at room temperatures.



It is known that this reaction can occur at room temperature and that the reaction rate significantly increases with increasing temperatures up to about 450°C. Because of the known reactivity of  $UH_3$  with air and water, one would expect that  $UH_3$  formation would be promoted under quiescent conditions. While  $UH_3$  does not react with strong alkaline solutions, it is not known whether  $UO_2$  (a basic oxide) possesses similar stabilizing capabilities.

Formation of  $UH_3$  on the metal surface could potentially increase uranium pyrophoricity by several mechanisms, namely:

1. Since  $\text{UH}_3$  is formed in an extremely finely divided state, this could be thought of as pyrophorically equivalent to converting small pieces of metal (at the surface) into a form having a relatively enormously increased surface to mass ratio. It is generally accepted that the pyrophoricity of  $\text{UH}_3$  is at least as great as that of metallic uranium of similar particle size.

2. Replacement of the protective and relatively non-porous  $\text{UO}_2$  film at the metal surface with a highly porous  $\text{UH}_3$  coating would be expected to increase the susceptibility of the surface metal to further attack by oxygen, hydrogen, or water.

A reaction mechanism similar to that outlined above has actually been substantiated by reported research on metallic calcium. Gibbs and Svec (J. Am. Chem. Soc. 75, 6052, 1953) investigated the reaction between calcium with water vapor obtaining, in sequence, unreacted calcium, a calcium hydride sub-scale, a thin uniform hydride layer, a mixed layer of hydride and oxide all overlaid with a thin calcium oxide layer in contact with the atmosphere. Absence of oxide in the hydride layers suggests, at least in the case of calcium that metal attack by water vapor may take place via a "hydrogen front" reaction at the metal surface. From the preceding, it is evident that at least partial circumstantial evidence exists to support the opinion that the mechanism causing increased uranium pyrophoricity may proceed along lines quite similar to the mechanism of calcium attack by water vapor as reported by Gibbs and Svec.

In essence, unusually high surface pyrophoricity of uranium is attributed to formation on the metal surface of a highly porous layer of  $\text{UH}_3$  protected by a thin unstable external coating of  $\text{UO}_2$ . The role played by stress and metallic contaminants in increasing uranium pyrophoricity is visualized as being quite similar to the roles played by these same factors in explaining normal corrosion of metal. Spontaneous combustion in uranium may be expected, according to this line of reasoning, following an initial highly exothermic local reaction of  $\text{UH}_3$  with air or water in which sufficient heat is rapidly created to raise the metal temperature in a local area to a point permitting triggering of similar reactions over adjacent surfaces of the metal (in which layers of  $\text{UH}_3$  may be alternated with relatively fire resistant sections of the metal covered by normal  $\text{UO}_2$  coatings). While it is difficult to conceive of surface reactions such as this being capable of spontaneously igniting massive pieces of metal, it should be noted that in the very few known cases in which spontaneous ignition of massive metal occurred at room temperatures, the metal involved is believed to have been made by powder metallurgical means, using uranium hydride as the starting material. It is suspected, therefore, that the degree of pyrophoricity of massive uranium is at least partially governed by the quantity of dissolved or occluded hydrogen and hydride present in the metal at the time of its initial preparation.

**RESEARCH APPROACHES FOR REDUCING URANIUM PYROPHORICITY**

Knowing that the pyrophoricity of metallic uranium appears to be increased when alloyed with such elements as tin, lead, and gold, it would be of interest to determine whether addition of any elements in trace quantities could be found which would be capable of significantly lowering uranium combustibility. As a precedent, it is of interest to note that the pyrophoricity of magnesium-aluminum alloys can be appreciably reduced by the addition of beryllium in amounts producing maximum effectiveness in concentrations of 0.001 to 0.003 percent. Perhaps significantly, it has also been established that the same concentration of beryllium is capable of affording maximum protection of magnesium-aluminum alloys against reaction with steam (J. Inst. Metals, 82, pp. 107 and 114, [1954]).

If the research program at the Argonne National Laboratory should confirm the suspicion that uranium pyrophoricity is promoted as a consequence of hydride formation, it would follow that uranium pyrophoricity could potentially be reduced to a minimum by provision of any mechanism tending to retard or prevent hydride formation. In this connection, it has been demonstrated on a laboratory scale that the rate of corrosion of aluminum by water at elevated temperatures can be significantly reduced if a suitable metal is present on the aluminum surface to serve as a local point for formation and release of hydrogen.

**URANIUM-WATER EXPLOSIONS**

Very few metals are capable of releasing more energy during water-metal reactions than can be theoretically obtained from uranium-water reactions. That explosive-water-metal reactions of any type very rarely occur suggests that the conditions necessary to achieve explosive reactions are quite narrow. Three experimental methods are known to have been used to obtain water-metal explosions under controlled conditions, namely:

1. Explosion of a detonator cap in a suspension of finely divided metal powder in water.
2. Pouring molten metal into water.
3. Extremely rapid heating of a metal wire immersed in water (by rapid discharge of a charged condenser).

All of the above methods suggest that explosive water-metal reactions may only occur under dynamic and perhaps transient conditions. Limited laboratory tests appear to have established that these explosive reactions are of exceedingly short duration (micro or millisecond range) and are self-limiting to the extent that the conditions necessary to initiate explosion are apparently disrupted after a relatively small amount of the metal has reacted with water.

It has previously been pointed out that a connection is thought to exist between factors tending to increase uranium pyrophoricity, factors involved in corrosion of uranium by water, and factors involved in water-metal explosions. It is important to note in this connection that all of the methods for producing water-metal explosion outlined in the preceding paragraph are capable of producing extremely high but momentary pressures at the water metal interface. While this is obviously true where detonator caps are used, it is less apparent but equally true when hot metal suddenly contacts water (or vice versa) as a consequence of transient momentum effects. Under the latter conditions, very rapid evolution of steam and hydrogen bubbles would be expected. But as such bubbles start to form, water must first be displaced and for this to occur a force (pressure) must exist in an amount proportional to the rate of bubble formation (and of the rate of water displacement). Since the momentary driving force tending to create bubbles (i.e., heat and chemical reaction) is extremely high, it follows that correspondingly extremely high pressures would momentarily exist before bubble formation starts. Following bubble formation, pressures would be expected to fall rapidly, in part due to decreased heat transfer through bubbles and mass action considerations.

Consider then the following possible sequence of events that may transpire during and immediately after the very brief period during which extremely high pressures exist at the water-metal interface preceding a water-metal explosion. From the limited available information, the following is suggested as a possible sequence of events that may transpire:

1. The rate of hydrogen formation under the assumed conditions would be roughly proportional to the temperature at the uranium surface. Due to the extremely high pressure, a strong force would exist tending to prevent hydrogen bubble formation, but the same force would also tend to materially increase the rate of absorption of hydrogen at the metal surface (as well as promoting formation of uranium hydride).
2. As bubbles form at the water-metal interface, rapid lowering of pressures in this area would be expected, as previously explained. But with the lowered pressure, literal spewing of very hot and very finely divided metallic uranium into the water may be expected in part due to thermal decomposition of  $\text{UH}_3$ .
3. Finally, the actual water-metal explosion is thought to involve an extremely violent, rapid reaction between water and the very hot and finely divided particles of metal and uranium hydride blown out from the metal surface.

If the above line of thinking regarding the mechanics that may be involved in a water-metal explosion is confirmed as a consequence of future research, it would be expected that one line of attack in preventing such explosions would be to attempt to minimize the rate of hydrogen solution and of hydride formation at the surface of metal exposed to water. Thus, one method of potentially minimizing the risks of en-

countering a water-metal explosion is identical in principle to the major indicated line of attack for preventing abnormally high pyrophoricity in uranium.

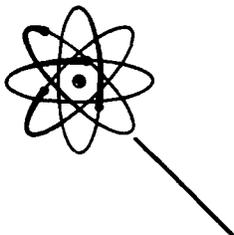
## CONCLUSIONS

There is need for further research to explain the causative factors for the vagaries that are occasionally encountered in the pyrophoricity of metallic uranium. The study of circumstantial evidence in numerous uranium fires investigated to date suggests that such vagaries are probably largely an aftermath of prior uranium-water reactions under conditions conducive to formation of uranium hydride. There are reasons for thinking that the mechanics for explaining water-metal explosions will be found to be quite similar to those explaining reasons for occasional existence of uranium metal having abnormally high pyrophoricity.

Detailed research into the mechanics involved during combustion of certain heavy metals has been undertaken by the Argonne National Laboratory under AEC contract. As a consequence of this research, it is hoped that means will be found for better evaluation and control of fire and explosion risks during uranium metal formation, fabrication, and ultimate use.

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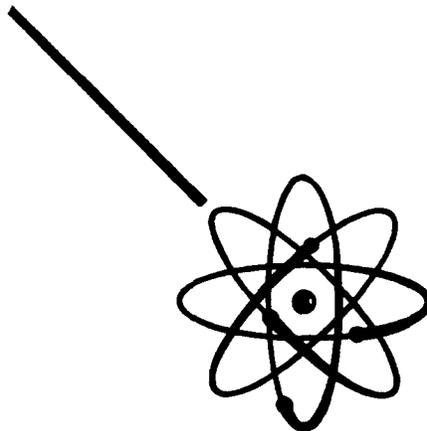
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## THE FIRE PROPERTIES OF METALLIC URANIUM

by R. B. Smith

In general, the fire properties of metallic uranium are quite similar to, but more pronounced than, those of magnesium of similar form. Under suitable conditions, uranium is capable of self-sustaining combustion in air, nitrogen, oxygen, and carbon dioxide. Investigators are agreed that the reaction at high temperatures between steam and uranium is much more vigorous than that between uranium and oxygen. While quite rare, severe spontaneous fires have been experienced during handling of fine moist powders under water as well as under inert gas (helium and argon). Burning uranium is capable of violently reacting with all commonly used fire extinguishants.

Self-sustaining combustion of uranium in air is accompanied by emission of very intense but localized heat and light. The rate of combustion may range from explosively violent, when finely divided metal powders are dispersed in a dust cloud, to very slow quiescent burning in the case of massive metal. Complete self-sustaining combustion of a one-inch diameter rod, for example, may require up to 24 hours before all of the metal is ultimately converted into the oxide. The metal burns without flame. The heat created during a fire has often been sufficient to burn holes through supporting thin sections of steel or cast iron. Once initiated, the rate of metal combustion is little affected by the porous oxides that are created.

That the pyrophoricity of metallic uranium can vary over an extremely wide range is attested to by the many vagaries experienced to date. For example, it is often possible to heat massive pieces of metallic uranium in air to temperatures up to the melting point without initiating oxidation at a rate sufficient to obtain self-sustaining combustion when the heat source is removed. By contrast, one case was reported in which a "yeast cake" size piece of uranium ignited spontaneously while resting on a block of dry ice!

Before further discussion of the vagaries in uranium pyrophoricity, it is important to point out that:

1. Massive uranium is normally incapable of self-sustaining combustion in air without the continued application of heat from external

sources. The occurrence of fires during rolling, extrusion, forging, storage, etc., is exceedingly rare even when manufacturing sheet uranium strip.

2. The frequency of fires that may be expected while metallic uranium is being sawed or while lathe or milling operations are being performed will vary with the type and rate of coolant application.

a. If no coolant is used, the chips or sawdust will often spontaneously ignite in air at the time of their creation.

b. If mineral oil coolants are liberally applied during machining operations, few if any spontaneous fires may be expected to occur in the chips.

c. If water (or a water soluble vegetable oil) is copiously applied as coolant, sporadic minor fires may be expected in the metal chips or sawdust at the time of their creation. Such a fire in a single lathe chip normally "burns itself out" after a few seconds duration but may on occasion spread to other chips in the immediate area. When this occurs in a small amount of lathe turnings, the fire may generally be easily extinguished by "dousing with coolant." A fire involving a large amount of chips on a lathe bed may be difficult or impossible to extinguish. While uranium chips, turnings, and sawdust do not normally spontaneously ignite during subsequent handling in air, literally thousands of spontaneous fires of this type have been suffered. Extinguishment or control of such fires is difficult or impossible where large accumulations of chips or turnings are involved. Controlled incineration or digestion by chemical means afford the safest methods for averting fire hazards from uranium chips and turnings. Where this is not possible, care should be taken to keep accumulations of metal chips at any one point to a minimum. The hazards of spontaneous ignition may be minimized by storage of chips under mineral oil although under these conditions gaseous hydrogen will be very slowly evolved. When chips are stored under water (or water soluble coolants), hydrogen evolution takes place somewhat more rapidly.

3. Finely divided metal powders created by any means are particularly pyrophoric. Grinder sludge, for example may spontaneously ignite while immersed in water for periods of a day or more. Dry uranium powder normally ignites spontaneously when exposed to air. If the metal powder is dispersed in air, spontaneous explosions may be expected. A detailed investigation into the hazards of uranium powder has been reported by Hartman (Bureau of Mines Report No. 3202).

#### VAGARIES IN URANIUM PYROPHORICITY

Unusually high pyrophoricity may be displayed in uranium either as a purely surface phenomenon or less frequently as a uniform character-

istic of all metal in the specimen. The following incidents will serve to illustrate some of the vagaries experienced to date:

1. Incidental to the manufacture of metallic uranium powder, some of the powder was slowly accumulated in a sump under perhaps 20 feet of sea water. Such accumulations are understood to have built up for periods of a month or so without incident following which, without prior warning, a vigorous reaction took place resulting in creation of a local "geyser." Following such eruptions, no further eventful happenings occurred until subsequent accumulations of metal powder permitted repetition of the preceding event.

2. Specimens of massive, low density (e.g., density of 15) metallic uranium are reported to have spontaneously ignited in air after having rested without incident for several days on a metal shelf.

3. The external jackets on several experimental uranium fuel elements failed after exposure in a water-cooled reactor. When the jackets of these fuel elements were removed by remote control means (they were highly radioactive), existence of powdered material not present when the elements were fabricated was noted. The powder spontaneously ignited when exposed to air.

4. Following the hot rolling of uranium billets into rods, the rods (roughly 1 inch in diameter) were placed on a straightening rack where they were permitted to cool to room temperature. Occasionally a rod would be encountered which not only failed to cool but instead would slowly increase in temperature until self-sustaining combustion was initiated and continued for approximately 24 hours until all of the metal had burned to the oxide. While numerous and simultaneous changes made during this period preclude pinpointing reasons for the subsequent significant decrease in the number of "pyrophoric" rods encountered, the presence of metal contaminants (particularly iron and copper) was suspected to be a major causative factor.

5. A large number of massive uranium pieces approximately one-half inch thick was unintentionally exposed to heat from an external fire of approximately one hour duration. The uranium ignited and continued to burn (without the application of heat from any external source) for almost a day. It should be pointed out that it is normally impossible to obtain self-sustaining combustion of massive uranium metal without continued application of external heat.

6. Prior to anticipated shipment, scrap uranium metal was incinerated to convert it into the oxide following which it was cooled to room temperature prior to packaging. When the "oxide" was received several days later at its destination, fire broke out in the packing material. Investigation revealed pieces of burning metal within the oxide.

7. Moist powdered uranium on steel trays was placed on cast iron, steam heated shelves of a vacuum dryer. After purging with an inert gas (argon) and application of vacuum, a spontaneous fire would occasionally occur of such intensity as to burn holes through both the supporting tray and the supporting cast iron grid.

8. A series of spontaneous fires had occurred in compacted uranium chips stored in closed-top 55-gallon metal drums under outside weather conditions. An investigator opened one such drum and, after noting that the drum contents were at room temperature and that everything appeared to be normal inside, was warned by an operator to "stand back." About 15 seconds later, the investigator witnessed a column of fire shoot up roughly 25 feet into the air and then immediately subside. Following this, it was found that the entire contents of the drum were incandescent.

9. Loose, dry uranium chips had been stored for about 6 months at room temperature in small (about 2 quarts) stainless steel containers having loose-fitting lids. On Saturday, a guard observed that in the area of these cans the temperature had risen to 105°F and called a supervisor who inspected each of the cans, noting nothing unusual, and left after correcting conditions which had led to previous overheating of the room. Temperature in this room over the weekend was at normal levels. Monday morning the supervisor again inspected the cans and this time found that many of them showed evidence of having been exposed to a high temperature. In part, this evidence consisted of holes that had been burned in the sides of the cans. Investigation revealed that while fire had unquestionably occurred in the uranium within the cans, only 15 percent of the metal (on an average) had been converted into the oxide.

10. During experimental research, it was found that two small batches of uranium powder appeared to have been rendered "nonpyrophoric" after treating the powder with carbon tetrachloride. When this was tried on a third and larger batch, a violent explosion occurred severely injuring the person conducting the experiments.

11. After removal of loose uranium chips from a nitric acid bath, small localized spontaneous fires have been occasionally encountered. When such fires occur in the open, they have on many occasions been extinguished by applying water with a hose. When fires of this type occur under confined conditions, they have been extinguished by flooding with CO<sub>2</sub> or in extreme cases with argon (although in rare cases even the latter fails to provide extinguishment of the blaze). On several occasions, localized fires in chips wet with nitric acid (handled in the open) have not only spread throughout the chips in the container, but have also been accompanied by explosions of sufficient violence as to seriously damage adjacent walls and ductwork.

12. Uranium powders (which are normally prepared by pyrolysis of uranium hydride) are usually highly pyrophoric. That the pyrophoricity of such uranium powder may vary over quite a wide range is fairly well recognized. (In this connection, it is of interest to note that catalytic iron prepared by hydrogen reduction of iron formate at elevated temperatures spontaneously ignites when suddenly exposed to air at room temperature.) Various patents have been issued for "stabiliza-

tion" of pyrophoric metal powders such as patents 2,677,668 and 2,677,669 issued to J. E. Ahlberg in May 1954.

13. It has previously been pointed out that spontaneous fires have been reported to have taken place while uranium powder or chips were submerged in water. Conversely, however, many fires involving incandescent uranium chips have been extinguished by immersion of the chips into water. Similarly, it is known that self-sustaining combustion of uranium can take place at elevated temperatures with  $\text{CO}_2$  but despite this fact operators have occasionally been successful in extinguishing uranium fires using  $\text{CO}_2$  extinguishers.

While the above list does not exhaust the types of vagaries experienced to date during handling of metallic uranium, it is perhaps sufficient to partially illustrate the basis for the following tentative conclusions which were reached after study of the available circumstantial evidence involved in hundreds of uranium fires:

1. Unusually high uranium pyrophoricity may be acquired as a consequence of factors associated with the initial preparation of the metal (this point has been confirmed by research in the case of metallic plutonium) or it may be acquired at a later period as a consequence of metal exposure to air and moisture under quiescent conditions.

2. The presence of certain metal contaminants or existence of the metal in a stressed condition (e.g., lathe chips) tends to increase the probability that a uranium specimen will acquire added pyrophoricity after initial formation of the metal.

3. Spontaneous ignition of uranium involves a rapid initial exothermic surface reaction with air or water. As contrasted to the better known cases of spontaneous ignition (e.g., oily rags), the spontaneous ignition of uranium does not appear to be preceded by any slow, gradual build-up of temperature.

4. Uranium pyrophoricity is strongly influenced by the nature of the surface oxide. Surface oxides acquired during metal exposure to dry air appear capable of very materially reducing (if not eliminating) the ability of a metal specimen to acquire added pyrophoricity. If other factors are equal, the degree of pyrophoricity increases with increasing surface to mass ratio of metal specimen.

5. In common with many other metals, uranium combustion in air is promoted by the presence of moisture in limited amounts.

6. Low density (e.g., apparent density of 15) specimens of uranium metal prepared by powder metallurgical means through decomposition of compressed uranium hydride are believed to have been involved in most if not all known fires in which massive pieces of metal spontaneously ignited while at room temperatures. It should be noted, however, that metal produced in vacuum casting operations or metal prepared by bomb reduction means will also occasionally display unusually high pyrophoricity particularly at the metal surface.

## THEORIES AS TO THE MECHANICS CONTRIBUTING TO URANIUM PYROPHORICITY

Any explanation as to the mechanics causing unusually high pyrophoricity in metallic uranium must take into consideration the conclusions detailed in the preceding section. Study of uranium fires experienced to date affords particularly strong circumstantial evidence on the following points:

1. Metal exposed to moisture under quiescent conditions (and preferably in the presence of air) appears to be almost invariably necessary before uranium can acquire unusually high pyrophoricity.

2. The probability that a particular uranium specimen will be unusually pyrophoric is greatly increased when the metal exists in a stressed condition and/or when it contains certain metal contaminants.

3. Unusually high pyrophoricity may exist either as a purely surface phenomenon or, less frequently, may be a property characteristic of all metal in the specimen.

The ability of moisture to affect metal pyrophoricity was dramatically displayed in the following incidents:

1. An AEC contractor who handled uranium chips in dried air very rarely encountered any incidence of spontaneous fires. When the air dryer failed, a relatively large number of spontaneous fires was experienced. When the air dryer was repaired and replaced in service, the incidence of such fires again fell to practically zero.

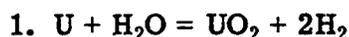
2. The following incident provides unique and spectacular evidence that pyrophoricity of metals other than uranium may also be markedly influenced by exposure to moisture. Trouble was experienced in getting a reaction of gaseous zirconium tetrachloride with metallic magnesium to go to completion. When the furnace was opened, a thin surface crust was found which was thought to consist of a mixture of zirconium, magnesium, and magnesium chloride. A piece of this crust, roughly one square foot in area removed for inspection and test, appeared to be inert (i.e., no sparks) when scratched with a file and hit with a hammer. The piece was next placed under water for the purpose of leaching out the  $MgCl_2$ . This operation was repeated three or four times without any unusual occurrence. During the last attempted extraction and without prior evidence of any reaction, a violent explosion occurred (while the piece was under water) which shattered the laboratory bench, blew out a window some 20 feet away, and seriously injured the investigator. During subsequent cleanup, particles of metal found in adjacent plaster walls spontaneously ignited when the surface layers were touched with a brush. This incident is considered significant in strengthening the circumstantial evidence that:

- a. Under suitable exposure conditions, metal pyrophoricity can be materially increased by exposure to water.

b. Spontaneous metal fires need not be preceded by any slow buildup of heat.

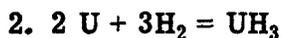
c. A connection probably exists between factors causing added metal pyrophoricity and factors involved in water-metal explosions.

If one accepts the premise that uranium pyrophoricity can be increased as a consequence of metal exposure to water under suitable conditions, the following known reactions must be considered:

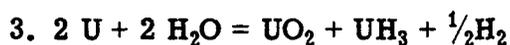


There is no evidence that this exothermic reaction has ever taken place at room temperature at a rate sufficiently rapid to cause significant heating of the metal. It is known that at room temperature hydrogen-saturated water corrodes uranium much more rapidly than water saturated with oxygen. Both conditions may simultaneously exist when the metal is exposed to quiescent moist air, thus introducing electrolytic factors, the net affect of which has not been extensively studied. Only one of the many investigators of the reaction reported (in 1943) that the  $UO_2$  produced was pyrophoric (see pp. 303 and 309, Vol. VIII-5, of the National Nuclear Energy Series).

While additional heat from hydrogen combustion may increase the rate at which uranium burns in air, this cannot account for the uranium fires that have taken place in the absence of air. Thus, there is little, if any, evidence that the subject reaction in itself can promote unusually high metal pyrophoricity.



This reaction has also been extensively investigated and could conceivably occur as a consequence of hydrogen evolved in reaction 1, above. The  $UH_3$  is brittle, extremely finely divided and highly reactive with both air and water (although this reactivity is quite erratic where small amounts of  $UH_3$  are involved). There is little reason to believe this reaction could occur to any significant degree except under quiescent conditions tending to prevent immediate conversion of any  $UH_3$  formed to  $UO_2$ . Once initiated, the uranium-hydrogen reaction is quite rapid even at room temperatures.



It is known that this reaction can occur at room temperature and that the reaction rate significantly increases with increasing temperatures up to about 450°C. Because of the known reactivity of  $UH_3$  with air and water, one would expect that  $UH_3$  formation would be promoted under quiescent conditions. While  $UH_3$  does not react with strong alkaline solutions, it is not known whether  $UO_2$  (a basic oxide) possesses similar stabilizing capabilities.

Formation of  $UH_3$  on the metal surface could potentially increase uranium pyrophoricity by several mechanisms, namely:

1. Since  $\text{UH}_3$  is formed in an extremely finely divided state, this could be thought of as pyrophorically equivalent to converting small pieces of metal (at the surface) into a form having a relatively enormously increased surface to mass ratio. It is generally accepted that the pyrophoricity of  $\text{UH}_3$  is at least as great as that of metallic uranium of similar particle size.

2. Replacement of the protective and relatively non-porous  $\text{UO}_2$  film at the metal surface with a highly porous  $\text{UH}_3$  coating would be expected to increase the susceptibility of the surface metal to further attack by oxygen, hydrogen, or water.

A reaction mechanism similar to that outlined above has actually been substantiated by reported research on metallic calcium. Gibbs and Svec (J. Am. Chem. Soc. 75, 6052, 1953) investigated the reaction between calcium with water vapor obtaining, in sequence, unreacted calcium, a calcium hydride sub-scale, a thin uniform hydride layer, a mixed layer of hydride and oxide all overlaid with a thin calcium oxide layer in contact with the atmosphere. Absence of oxide in the hydride layers suggests, at least in the case of calcium that metal attack by water vapor may take place via a "hydrogen front" reaction at the metal surface. From the preceding, it is evident that at least partial circumstantial evidence exists to support the opinion that the mechanism causing increased uranium pyrophoricity may proceed along lines quite similar to the mechanism of calcium attack by water vapor as reported by Gibbs and Svec.

In essence, unusually high surface pyrophoricity of uranium is attributed to formation on the metal surface of a highly porous layer of  $\text{UH}_3$  protected by a thin unstable external coating of  $\text{UO}_2$ . The role played by stress and metallic contaminants in increasing uranium pyrophoricity is visualized as being quite similar to the roles played by these same factors in explaining normal corrosion of metal. Spontaneous combustion in uranium may be expected, according to this line of reasoning, following an initial highly exothermic local reaction of  $\text{UH}_3$  with air or water in which sufficient heat is rapidly created to raise the metal temperature in a local area to a point permitting triggering of similar reactions over adjacent surfaces of the metal (in which layers of  $\text{UH}_3$  may be alternated with relatively fire resistant sections of the metal covered by normal  $\text{UO}_2$  coatings). While it is difficult to conceive of surface reactions such as this being capable of spontaneously igniting massive pieces of metal, it should be noted that in the very few known cases in which spontaneous ignition of massive metal occurred at room temperatures, the metal involved is believed to have been made by powder metallurgical means, using uranium hydride as the starting material. It is suspected, therefore, that the degree of pyrophoricity of massive uranium is at least partially governed by the quantity of dissolved or occluded hydrogen and hydride present in the metal at the time of its initial preparation.

## RESEARCH APPROACHES FOR REDUCING URANIUM PYROPHORICITY

Knowing that the pyrophoricity of metallic uranium appears to be increased when alloyed with such elements as tin, lead, and gold, it would be of interest to determine whether addition of any elements in trace quantities could be found which would be capable of significantly lowering uranium combustibility. As a precedent, it is of interest to note that the pyrophoricity of magnesium-aluminum alloys can be appreciably reduced by the addition of beryllium in amounts producing maximum effectiveness in concentrations of 0.001 to 0.003 percent. Perhaps significantly, it has also been established that the same concentration of beryllium is capable of affording maximum protection of magnesium-aluminum alloys against reaction with steam (J. Inst. Metals, 82, pp. 107 and 114, [1954]).

If the research program at the Argonne National Laboratory should confirm the suspicion that uranium pyrophoricity is promoted as a consequence of hydride formation, it would follow that uranium pyrophoricity could potentially be reduced to a minimum by provision of any mechanism tending to retard or prevent hydride formation. In this connection, it has been demonstrated on a laboratory scale that the rate of corrosion of aluminum by water at elevated temperatures can be significantly reduced if a suitable metal is present on the aluminum surface to serve as a local point for formation and release of hydrogen.

## URANIUM-WATER EXPLOSIONS

Very few metals are capable of releasing more energy during water-metal reactions than can be theoretically obtained from uranium-water reactions. That explosive-water-metal reactions of any type very rarely occur suggests that the conditions necessary to achieve explosive reactions are quite narrow. Three experimental methods are known to have been used to obtain water-metal explosions under controlled conditions, namely:

1. Explosion of a detonator cap in a suspension of finely divided metal powder in water.
2. Pouring molten metal into water.
3. Extremely rapid heating of a metal wire immersed in water (by rapid discharge of a charged condenser).

All of the above methods suggest that explosive water-metal reactions may only occur under dynamic and perhaps transient conditions. Limited laboratory tests appear to have established that these explosive reactions are of exceedingly short duration (micro or millisecond range) and are self-limiting to the extent that the conditions necessary to initiate explosion are apparently disrupted after a relatively small amount of the metal has reacted with water.

It has previously been pointed out that a connection is thought to exist between factors tending to increase uranium pyrophoricity, factors involved in corrosion of uranium by water, and factors involved in water-metal explosions. It is important to note in this connection that all of the methods for producing water-metal explosion outlined in the preceding paragraph are capable of producing extremely high but momentary pressures at the water metal interface. While this is obviously true where detonator caps are used, it is less apparent but equally true when hot metal suddenly contacts water (or vice versa) as a consequence of transient momentum effects. Under the latter conditions, very rapid evolution of steam and hydrogen bubbles would be expected. But as such bubbles start to form, water must first be displaced and for this to occur a force (pressure) must exist in an amount proportional to the rate of bubble formation (and of the rate of water displacement). Since the momentary driving force tending to create bubbles (i.e., heat and chemical reaction) is extremely high, it follows that correspondingly extremely high pressures would momentarily exist before bubble formation starts. Following bubble formation, pressures would be expected to fall rapidly, in part due to decreased heat transfer through bubbles and mass action considerations.

Consider then the following possible sequence of events that may transpire during and immediately after the very brief period during which extremely high pressures exist at the water-metal interface preceding a water-metal explosion. From the limited available information, the following is suggested as a possible sequence of events that may transpire:

1. The rate of hydrogen formation under the assumed conditions would be roughly proportional to the temperature at the uranium surface. Due to the extremely high pressure, a strong force would exist tending to prevent hydrogen bubble formation, but the same force would also tend to materially increase the rate of absorption of hydrogen at the metal surface (as well as promoting formation of uranium hydride).

2. As bubbles form at the water-metal interface, rapid lowering of pressures in this area would be expected, as previously explained. But with the lowered pressure, literal spewing of very hot and very finely divided metallic uranium into the water may be expected in part due to thermal decomposition of  $\text{UH}_3$ .

3. Finally, the actual water-metal explosion is thought to involve an extremely violent, rapid reaction between water and the very hot and finely divided particles of metal and uranium hydride blown out from the metal surface.

If the above line of thinking regarding the mechanics that may be involved in a water-metal explosion is confirmed as a consequence of future research, it would be expected that one line of attack in preventing such explosions would be to attempt to minimize the rate of hydrogen solution and of hydride formation at the surface of metal exposed to water. Thus, one method of potentially minimizing the risks of en-

countering a water-metal explosion is identical in principle to the major indicated line of attack for preventing abnormally high pyrophoricity in uranium.

## CONCLUSIONS

There is need for further research to explain the causative factors for the vagaries that are occasionally encountered in the pyrophoricity of metallic uranium. The study of circumstantial evidence in numerous uranium fires investigated to date suggests that such vagaries are probably largely an aftermath of prior uranium-water reactions under conditions conducive to formation of uranium hydride. There are reasons for thinking that the mechanics for explaining water-metal explosions will be found to be quite similar to those explaining reasons for occasional existence of uranium metal having abnormally high pyrophoricity.

Detailed research into the mechanics involved during combustion of certain heavy metals has been undertaken by the Argonne National Laboratory under AEC contract. As a consequence of this research, it is hoped that means will be found for better evaluation and control of fire and explosion risks during uranium metal formation, fabrication, and ultimate use.

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