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The Use of (U, Pu)N Fuel with a Sodium Bond:
A Summary of Existing Data

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IN NUCLEAR SCIENCE ABSTRACTS

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The Use of (U, Pu)N Fuel with a Sodium Bond:
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by

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THE USE OF (U,Pu)N FUEL WITH A SODIUM BOND:

A SUMMARY OF EXISTING DATA

ABSTRACT

The pertinent literature is reviewed for an experimental study of the nuclear fuel (uranium, plutonium) mononitride bonded with sodium to a suitable clad. Sections on the fuel's physical properties, mechanical properties, compatibility, and resistance to reactor radiation are followed by a discussion on outstanding problems that still exist.

INTRODUCTION

The available literature on the use of (U,Pu)N fuel with a sodium bond is reviewed with emphasis on the last few years because almost all of the pertinent information has appeared during that period. The use of the nitride is typically envisioned as a solid fuel with a Pu:²³⁸U atomic ratio of 1:4. The nitride is currently considered as a possible, promising alternative to a fuel of carbide or oxide as are certain other ceramics such as uranium monosulfide or monophosphide.

PHYSICAL PROPERTIES OF THE FUEL

Compositions, Crystal Structures, and Stabilities

The most stable of the uranium nitrides and the only one considered for use as a fuel is the mononitride which has 5.56 wt. % N. Its vapor pressure, due to decomposition to nitrogen gas, is 10^{-6} atm at 1750°C,¹ 10^{-3} atm at 2250°C,¹ and 2.5 atm at its melting point of 2850°C.² The composition range of uranium mononitride is believed to be fairly small.³ Uranium sesquinitride is comparatively unstable as shown by its decomposition to the mononitride at 1520°, 1345°, and 780°C at nitrogen pressures of 5, 1, and 10^{-4} atm, respectively.³ Uranium dinitride only exists at still higher nitrogen pressures in solid solution with

the sesquinitride.⁴ The crystal structure of the mononitride is face-centered cubic with $a_0 = 4.880 \text{ \AA}$. That of the sesquinitride is body-centered cubic of a distorted fluorite structure with $a_0 = 10.678 \text{ \AA}$ and that of the dinitride is body-centered cubic of the ideal fluorite type with $a_0 = 5.31 \text{ \AA}$.⁵

Plutonium mononitride, the only stable nitride of plutonium, has an immeasurably small composition range. The pressure of nitrogen from its decomposition is 10^{-2} atm at 2300°C.⁶ Its stability is comparable to that of uranium mononitride at reactor temperatures. Its crystal structure is face-centered cubic with $a_0 = 4.905 \text{ \AA}$.⁷ It is isomorphous with uranium mononitride and forms solid solutions with it. The standard free energies of formation of uranium and plutonium mononitrides have been estimated for Table I.

Table I

Free Energies of the Fuel Mononitrides		
Compound	ΔF_f^0 (kcal)	Sources of Data
	at 1000°K	
UN	-49	8, 9, 1, 10
PuN	-53	11, 8, 12, 6

Mononitride pellets are ordinarily made at the Los Alamos Scientific Laboratory and elsewhere by pressing the powder accompanied by heat treatments.

The stoichiometric mononitride can thus be prepared more readily than the similar monocarbide, for example.^{13,14,15} However, as with the carbide, the nitride will have significant contaminants, notably oxygen as well as carbon. Small amounts of oxygen may be accommodated as uranium monoxide in solid solution in uranium mononitride up to 1 or 2 mole %.^{16a} The figure is "a few mole percent" for plutonium monoxide in plutonium mononitride.^{16b} Appreciably greater amounts of monoxide may be held in solid solution if the mononitride contains sizable amounts of monocarbide. Uranium mononitride and monocarbide have complete solid solubility for each other.¹⁷

The mononitride pellets may have some nonstoichiometry at the surface for at least two reasons: some uranium metal may have been produced at the surface due to local overheating and evolution of nitrogen during heat treatment¹⁸ and the pickup of oxygen during pressing or subsequent heat treatment of the pellet gives uranium monoxide and/or dioxide. A by-product of this oxidation is uranium sesquinitride.^{19a,b,c} Unless the surface area is cleaned of these impurities, for example by machining, the compatibility with metal claddings will be impaired.

Density, Expansion Coefficient, Heat Capacity, Thermal Conductivity, and Resistivity

A figure for the readily attainable density (95% of theoretical) of uranium mononitride is 13.6 g/cm³ at room temperature.²⁰ The nitride thus has a slightly higher uranium density than the carbide. Its linear thermal expansion coefficient is $9.9 \times 10^{-6} (\text{°C})^{-1}$ in the temperature range 20°-1600°C.¹⁴ Its molar heat capacity is given by $C_p (\text{cal/°C}) = 13.32 + 1.19 \times 10^{-3} T - 2.10 \times 10^{-5} T^2$, where T (°K) is between 273° and 1422°K.²⁰ The thermal conductivity for fully dense specimens increases from 0.21 watts/cm °C at 850°C to 0.26 watts/cm °C at 1200°C.²¹ These values are higher than those of the monocarbide above 1000°C, worse below 1000°C. The mononitride's electrical resistivity increases from 170 microhm-cm at room temperature to 210 microhm-cm at 1200°C.²¹

PHYSICAL PROPERTIES OF THE SODIUM

The physical and other properties of sodium are comparatively well known;^{22,23,24} hence no review of them will be given.

MECHANICAL PROPERTIES OF THE FUEL

Tensile properties have been determined on uranium mononitride that had been isostatically hot pressed to a fully dense material.¹⁴ The values (at 25°C) are: Young's modulus = 31×10^6 psi and shear modulus = 14×10^6 psi from which Poisson's ratio is 0.1 approximately. The bulk modulus has been given as 25×10^6 psi by Speidel.²⁵ Compressive creep has been determined for uranium mononitride at 1100°-1350°C.²⁶ The values are less than those for uranium monocarbide. The creep rate ($\dot{\epsilon}$) can be represented by

$\dot{\epsilon} = 2.75 \times 10^5 \exp(-62,000/RT)$ at 6,000 psi stress and

$\dot{\epsilon} = 6.9 \times 10^5 \exp(-60,800/RT)$ at 8,000 psi stress

The data give a value of $n = 4.15$ for $\dot{\epsilon} = A\sigma^n$, where σ is the stress. A typical creep rate at 1200°C and 8,000 psi stress is $8 \times 10^{-4} \text{ hr}^{-1}$. Hardness values have been determined for the mononitride at various temperatures.¹⁴ The DPH (diamond pyramid hardness) value varies from 499 at 25°C to 53 at 1400°C. The hardness drops rather abruptly above 1100°C. To quote Speidel and Keller, "Although hardness may not be unequivocally interpreted as a measure of mechanical strength, the retention by UN of about 80 per cent of its room-temperature hardness at 1100°C suggests that a large percentage of its mechanical strength will also be retained at that temperature."¹⁴

Uranium nitride pellets of 1/2 in. x 1/2 in. have been cycled between about 1100°C and room temperature over a period of a few minutes for 18 cycles.²⁷ Neither they nor similar pellets of uranium monocarbide or uranium dioxide showed any changes such as cracking or spalling due to thermal stress. However, the cracking or fracture of these ceramics is sometimes attributed to thermal shock during tests on radiation resistance or mechanical properties. It is probable that uranium nitride and carbide are better than the dioxide in resistance to thermal shock, principally on the basis of thermal conductivity.

COMPATIBILITY

With Air, Gases, and Non-Metallic Liquids

As usually prepared, uranium mononitride is less subject than uranium monocarbide to attack by ordinary air.²⁸ The mononitride's attackability is dependent on its purity, especially that at the surface. The attack by air is probably due mostly to the moisture content. This is suggested in some exposures of fully dense mononitride to one atmosphere of oxygen, water-saturated air, or carbon dioxide at 300°-700°C.⁴ The attack in each case is first order and produces U_3O_8 , which adheres to produce a gain in weight but is not protective. The rate constants (mg/cm^2 sec) at 300°C are 1.1×10^{-4} for oxygen, 1.1×10^{-3} for water-saturated air, and 1.2×10^{-3} for carbon dioxide. Hence, it is inferred that the oxygen in the air is not the rate-controlling corrosive agent; rather it is the moisture of the air. The rate of reaction of nitrogen with the mononitride appears not to have been studied but it is assumed to be comparatively slow below 600°C. Hydrogen gives no reaction with the nitride up to 800°C.²⁵

Only stoichiometric, fully dense uranium mononitride of all of the uranium-nitride preparations shows any resistance to hot water.⁴ This nitride develops a film of UO_2 , which gives a loss in weight as it flakes off. In boiling water at 100°C, the weight loss is $1.3 mg/cm^2$ in 1047 hr. The weight loss is $560 mg/cm^2$ in 300°C water for 48 hr. The attack of stoichiometric, hypo-, or hyperstoichiometric uranium nitride in Santowax R at 350°C is slight. A film of UO_2 is produced. The attack is believed to be due to the moisture content.⁴ Any of these nitrides may be cleaned of NaK without change by using butyl and methyl alcohols.⁴

Plutonium mononitride is apparently less resistant to water than is the uranium compound. Pardue *et al.* report: "No attack on PuN by cold water was observed in 24 hr, but in boiling water, conversion to hydrated plutonium oxides was rapid, occurring within 10 min."¹⁵ Of plutonium nitride's reactions with stock reagents at room temperature they say: "In order of decreasing attack, the acids studied were: HNO_3 , HCl , H_3PO_4 , H_2SO_4 , and HF . Dissolution in HNO_3 was complete within 1-1/2 hr, whereas in HF ,

the material exhibited only crumbling after 24 hr." They also report no visible effect from the use of butyl alcohol in dissolving sodium or lithium from plutonium nitride.

With Liquid Metals

Samples of stoichiometric, hypo-, and hyperstoichiometric uranium nitride have been exposed to 820°C NaK for 1000 hr without detectable effect.⁴ Nor is uranium mononitride severely attacked by 480°C NaK in 1500 hr.¹⁴ Plutonium nitride has been exposed to sodium and lithium of commercial grade, these liquid metals being gettered during test by zirconium chips or chips of a titanium-zirconium alloy.¹⁵ The results "are of a preliminary nature, but suggest that PuN is compatible with sodium and lithium at temperatures up to at least 800°C" for 336 hr. "In the temperature range 950 to 1000°C, however, PuN disintegrated in both sodium and lithium" in about the same time.

With Directly Bonded Clads

A few papers have appeared on the nitride's compatibility in direct contact with the cladding. Iron and Type 304 stainless steel have been recognized as among the most compatible of clads for uranium mononitride at temperatures of 1000°C²⁹ or slightly higher.^{19a} Katz has found no attack for either clad in 500 hr at 1000°C.²⁹ What little attack Price *et al.* have found with iron in 3000 hr at 1000°C has been attributed to the oxygen impurity.^{19a} Their observations with Type 304 stainless steel at 1000°-1300°C for over 1000 hr have indicated only a slight attack due to impurities. They found the formation of less than 1 mil of a reaction layer containing only Cr_2N as a *bona fide* reaction product. Pardue's data on these two clads with plutonium mononitride have shown no reaction in 100 hr at 1300°C.¹⁵ Nor have these clads shown a reaction with slightly hypostoichiometric plutonium mononitride in 1000 hr at 820°C.³⁰ Katz's data have indicated poorer compatibility of these two clads with his uranium monocarbide.²⁹ Presumably other iron-based alloys, e.g., Type 316 stainless steel, also might be considered for compatibility with uranium or plutonium mononitride.

Price *et al.* say that chromium probably does not react with stoichiometric uranium mononitride in

1000 hr at 1000°C.^{19a} In their studies UO_2 and about 2 mils of Cr_2N "formed probably as the result of the presence of oxygen in the chromium and of U_2N_3 in the UN, respectively." Katz also has observed only a slight reaction of uranium mononitride with chromium in 500 hr at 1000°C.²⁹ The Battelle group has observed no reaction between chromium and slightly hypostoichiometric plutonium mononitride in 1000 hr at 820°C.³⁰

Nickel, the third largest component of the 300-series stainless steels, is attacked comparatively rapidly by either uranium or plutonium mononitride.^{19a,15,30} The extensive data of Price *et al.* show the principal reaction product with uranium nitride to be a layer of UNi_5 sometimes accompanied by a detectable layer of U_2N_3 .^{19a} The principal corrosion reaction is of the type

Their data show about an 8-mil reaction layer in 1132 hr at 1000°C. Their results with nickel are qualitatively supported by those of Katz²⁹ and of Anselin *et al.*³¹ Slightly hypostoichiometric plutonium nitride has shown a one-third mil reaction layer with nickel, but none with Inconel, in 1000 hr at 820°C.³⁰ Inconel also has shown no reaction with plutonium nitride in 100 hr at 1300°C. Apparently Inconel's alloying elements impede the attack in spite of the alloy's being three-fourths nickel.

In tests of 500 hr at 1000°C, Katz has found molybdenum to give a reaction layer of 0.1 mil with uranium nitride and niobium to give one twice as thick.²⁹ Tungsten has been observed to undergo no attack in 2-3 hr at 2800°C with this nitride.²⁰ Pardue *et al.* have observed molybdenum, niobium, tungsten, and tantalum in contact with plutonium nitride.¹⁵ They have found no reaction with any of these clads in 50 hr at 1288°C, but oxide impurities became evident in 200 hr at 1371°C or 96 hr at 1650°C. These four clads may be considered among the most resistant to nitride fuels, but iron is better at reactor temperatures than molybdenum or niobium.

Katz has found vanadium and zirconium to be somewhat worse than the above refractory metals in his tests with uranium nitride for 500 hr at 1000°C.²⁹ The reaction layer is 0.5 mil with vanadium and

0.7 mil with zirconium. Zirconium nitride and an unidentified metallic phase are formed with the latter clad. Price *et al.* have found zirconium and Zircaloy-2 to behave very similarly toward uranium nitride.^{19a} The reaction products are $(\text{Zr,U})\text{N}$ and a metallic U-Zr solution, i.e., the corrosion reaction is of the following type:

The total thickness of these reaction layers is about 0.5 mil in 5000 hr at 700°C. A clad of titanium or thorium appears to give the same type of reaction as zirconium, but at a much greater rate. The reaction layer with either of these clads is greater than 5 mils in 500 hr at 1000°C.²⁹

Aluminum has been shown to have no compatibility with uranium nitride above 500° or 600°C.^{32,29,19a} For example, Price *et al.* have found the thickness of the reaction layer to be 67 mils in 100 hr at 600°C. The reaction layer consists of UAl_3 ,^{29,19a} probably other U-Al compounds,^{29a} and possibly AlN .^{19a}

RADIATION RESISTANCE

Nuclear Properties of the Fuel

The principal parasitic reaction of nitrogen with neutrons is the (n,p) reaction for which the cross section is about 0.0015 barn at 100-200 keV, which may be taken as representative of the average flux in a fast reactor.³³ This reaction also has strong resonances at slightly higher energies. There is also an appreciable (n,α) reaction in a fast flux. The total parasitic cross section is about 1.8 barns in a thermalized spectrum.³⁴ Although this removal of neutrons is tolerable regarding neutron economy, the reaction might possibly be a troublesome source of hydrogen (and helium) gas in the fuel element.

Burnup Experiments

Irradiations by R. L. Martin *et al.* indicate that the produced hydrogen is readily released from the uranium nitride at 500°-700°C, but it does not easily diffuse through Type 316 stainless steel cladding at these temperatures.¹⁸ Hence, the produced hydrogen is not a serious source of radiation damage to the fuel itself.

Most of the radiation damage in uranium nitride near room temperature is caused by fission fragments. Annealing experiments have shown that the volume of such a sample is reduced toward normal again by about 30% recovery at 150°C and another 40% between 540° and 640°C.³⁵ About 30% of the lattice damage still remains at 640°C. The damage that anneals out at 150°C appears to be associated with the increase in electrical resistivity that is caused by radiation.

Most of the fission products are retained in uranium nitride at reactor temperatures; however, radio-iodine and the rare-gas fission products are partly released to the surroundings. Melehan and Gates have studied the rare-gas release ratios from samples of UN_{1.01} having 92 to 96% of the theoretical density.³⁶ The release ratio is approximately constant at 10^{-3} for these samples up to 800°C, above which it rises to 10^{-1} near 1600°C and can be described in terms of diffusion theory. The diffusion coefficients for the rare gases between 1000° and 1600°C are $D = D_0 \exp(-Q/RT)$, where $D_0 = (3 \pm 1) \times 10^{-6} \text{ cm}^2/\text{sec}$ and $Q = 60 \pm 5 \text{ kcal/mole}$. The gas release from UC_{1.04} of 93% of the theoretical density is comparable to that of the above nitride.

Among the earliest reactor irradiations of uranium mononitride were those of Wullaert *et al.*³⁷ Bare and 316 stainless-steel-clad pellets were irradiated in a thermal flux to burnups up to 3.8 at. % of the uranium at temperatures as high as 615°C at the surface and 1260°C (centerline) in NaK-filled capsules. The cladding, when present, was 15 mils thick. The hydrogen produced by the (n,p) reaction was not detrimental at these temperatures. The pellets of 3.8% burnup showed about 25% increase in hardness and less than 0.6% release of fission gases. The NaK did not appreciably attack either the cladding or the unclad specimens. The unclad specimens broke into fragments during the irradiations, however. The clad specimens did not fracture. The fuel's average density decrease (percent/ 10^{20} fissions per cm^3) was about 2.5 at an average irradiation temperature of 100°C, 0.8 at 200°C, and 0.4 at 400°-600°C. The nitride pellet produced a change in about 0.25 mil of the adjacent cladding during the 148 days of the most severe test. The latter may have been due

nitriding, but was not considered serious in any event.

The thermal irradiations of R. L. Martin *et al.* used cylindrical uranium mononitride clad with Type 316 ELC stainless steel that was 30 mils thick on the diameter, 50 mils on the ends.¹⁸ Burnups were in the range of 4.6 to 6.3%, and surface temperatures were up to about 750°C. The diametral swelling varied from 1.3 to 4%. All of the fuel broke within the cladding. The hydrogen produced in the fuel was released from the fuel but partly retained in the cladding whenever the cladding remained sound. The fission gas released from hot-pressed fuel ranged up to 26%, but was much less for sintered or arc-cast fuel. Two of the twenty fuel specimens partly melted eutectically with the clad, apparently due to some uranium metal in the fuel. Half of the stainless steel clads developed cracks due to embrittlement from an unidentified cause. The cause of the latter was probably not fast-neutron damage because the operation temperature should have annealed out such damage. It might have been due partly to a precipitation of sigma phase for which there was some metallographic evidence. Alternatively, the external NaK may have embrittled the clad by a selective leaching, particularly from the grain boundaries. This view was supported by some voids found near the cracks during metallography. Embrittlement due to the production of helium bubbles within the clad from $^{10}\text{B}(n,\alpha)^7\text{Li}$ was considered possible, although their metallography did not support this view very well. Hydrogen embrittlement could not be discounted although it is uncommon with an austenitic stainless steel. Cracking on the cladding's inner surface might have been due to localized concentrations of certain fission products like iodine and cerium. The cracking was regarded as a serious problem, but the performance of the uranium nitride itself was satisfactory. No fuel-clad chemical reaction was observed with the exception of the eutectic melting of the clad and supposedly uranium metal in the fuel.

Some interesting results have been obtained with UN - 20% PuN that was clad directly with Type 304 stainless steel and exposed in a thermal flux.³⁸⁻⁴² The second capsule underwent 2.5% total burnup and

developed a 4-8 mil, fuel-clad reaction zone with that area at about 600°C. The third capsule with 7.4% burnup showed much the same reaction zone. These two capsules showed considerable migration of the plutonium from the center of each pellet, the center becoming largely metallic fission products. Some UO_2 phase also appeared closely surrounding this central region. Further reports on these capsules will doubtless give more information on the nature of the fuel-clad reaction.

The thermal conductivity of uranium mononitride is reported to be halved by irradiations in a thermal flux to 2.1×10^{21} fissions/cm³, the centerline and surface temperatures being 1320° and 760°C, respectively.⁴³ A similar effect has been observed with uranium dioxide⁴⁴ and might exist with the monocarbide as well.

OUTSTANDING PROBLEMS WITH NITRIDE FUELS

Small amounts of contaminant oxygen have often been responsible for what little attack of the cladding has previously been observed. The stringent exclusion of contaminant oxygen and moisture must be a prime goal of any tests of compatibility. Perhaps an innocuous anti-oxidant such as uranium monoxide should be dissolved in the (U,Pu)N fuel.

There is no indication that plutonium nitride is less compatible with clads out-of-pile than uranium mononitride. There has been considerable out-of-pile testing of the compatibility of mononitride in direct contact with the cladding. The iron-based alloys appear very promising as do molybdenum, niobium, and perhaps a few other feasible materials. Data with sodium bonding should be obtained with these experimental clads. Nickel has been shown to be incompatible with nitride fuel, but some of its alloys, e.g., Hastelloy X, might be tested because of their other good properties and high alloy content. All of the above alloys should have good high-temperature strength and resistance to air, and other desirable characteristics such as resistance to radiation and good resistance to oxygen in sodium.

Tests are under way at the Battelle Memorial Institute with both sodium-bonded and pressure-contacted UN - 20% PuN, pure UN, and pure PuN in clads of various test materials.^{43,45-52} The thickness of

their sodium gap is 39 mils or more⁴⁶ in contrast to our planned value of about 7.5 mils. According to the most recent information, their test clads are Type 304 stainless steel (ordinary, ORNL modified, and extra low carbon content), Type 316 stainless steel, niobium - 1 wt. % zirconium, and molybdenum. The last three of these test clads are similar to ours. Their tests are for various combinations of 1,000-10,000 hr at 800°-1200°C. The different test conditions of our initial tests, e.g., 2000 hr at 750°C, will make a direct comparison of results difficult; however, there may be a meaningful comparison of their results with ours regarding the order of compatibility. For example, is the compatibility with UN - 20% PuN in the order 316 stainless steel > Mo(-0.5 Ti) > Nb - 1 Zr as might be inferred from the data of Katz?²⁹ Our tests should eventually be for at least 10,000 hr at the fuel-clad temperature expected in a reactor. This temperature is now thought to be about 700°C. Sodium-bonded combinations are now also being studied at the Pacific Northwest Laboratory, but details are lacking.

Among the more disquieting results to appear in the literature on nitride fuels are two pertaining to radiation resistance. The first is the strong decrease in the thermal conductivity of uranium mononitride under reactor irradiation.⁴³ This effect could result in the power rating of the nitride fuel being reduced, as is the case with the dioxide⁴⁴ and perhaps the monocarbide as well.

The second serious problem is the embrittlement under irradiation of Type 316 stainless steel observed by Martin *et al.* with an integrated thermal neutron flux of about 8.5×10^{20} neutrons/cm².¹⁸ However, this was not observed by Wullaert *et al.* under less stringent conditions.³⁷ Martin *et al.* listed possible explanations for their observed embrittlement. It is possible, of course, that the embrittlement would be less serious with other clads, even other ferrous alloys.

The radiation-induced swelling of uranium mononitride appears to be comparable at 3.8% burnup and temperatures above 400°C³⁷ to that of uranium dioxide.⁵³ Presumably such expansion could be accommodated with an appropriate design of the cladding.

No serious chemical reaction has occurred between a clad of Type 304 or 316 stainless steel and the mononitride when the reactor irradiation has been performed without oxygen reaching the fuel. Obviously more data are needed without the interference of oxygen. All of the irradiation data have been obtained with a thermal flux; however, data are needed in a fast flux such as that for the projected nitride fuel. More realistic effects would thus be obtained for fast-neutron damage and helium buildup in the cladding, for example. An integrated fast flux of about 6×10^{22} neutrons/cm² would be required to test a fuel piece to 10% burn-up if the fuel was (80% ²³⁵U - 20% Pu)N.

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