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**PRELIMINARY EVALUATION OF A PROPOSED
FUEL MATERIAL FOR HIGH TEMPERATURE REACTORS**

MASTER

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APPLIED CHEMISTRY RESEARCH**

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ABSTRACT

Results are reported for some preliminary experiments to determine the stability of solid solutions of UO_2 and ThO_2 in air at temperatures of 2500°F and above. The results are compared with those obtained by workers at Argonne National Laboratories during development work on the Borax IV experimental breeder reactor. It is concluded that further evaluation of the system at temperatures above 2500°F is required.

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PRELIMINARY EVALUATION OF A PROPOSED FUEL MATERIAL
FOR HIGH TEMPERATURE REACTORS

By

E.F. Juenke

INTRODUCTION

Recently some interest has been expressed in the system $\text{ThO}_2\text{-UO}_2$ both from the standpoint of a fuel additive for present ceramic fuel elements now under development and as an advanced fuel element for high temperature fast reactors.

Consequently, a program was undertaken to determine the high temperature stability of solid solutions of $\text{UO}_2\text{-ThO}_2$. This report summarizes the results obtained to date in a cursory evaluation conducted by the author and compares these results with those obtained by J.H. Handwerk in studies conducted at Argonne National Laboratory. Further investigations are indicated.

SUMMARY OF STUDIES CONDUCTED AT ANL⁽¹⁾

Preliminary development work on the preparation of $\text{ThO}_2\text{-UO}_2$ fuel elements for the Borax IV experimental breeder reactor has shown the following:

- (1) $\text{ThO}_2\text{-UO}_2$ form a continuous solid solution of cubic fluorite structure.
- (2) Additions of ThO_2 stabilize UO_2 in air up to 60 w/o UO_2 . Above this percentage, X-ray analysis shows the presence of a second phase identified as U_3O_8 .
- (3) The O/U ratio for the stable solid solution is 2.0 - 2.3.
- (4) Compositions containing up to 30 w/o UO_2 , prepared by reacting U_3O_8 and ThO_2 in air at 1750°C , show no change in dimensions or weight after reheating at 1400°C for 96 hours in air.
- (5) Irradiation testing of a 97.5 w/o ThO_2 - 2.5 w/o UO_2 to a 0.94 atom % burn-up of total metal atoms present shows no measurable dimensional changes but some microscopic cracking.
- (6) Fueled pellets of 93.6 w/o ThO_2 - 6.4 w/o UO_2 , fabricated by sintering U_3O_8 and ThO_2 at $1700^\circ\text{-}1750^\circ\text{C}$ for 2 hrs. in an over-all cycle of 16 hrs., attained 97% theoretical density.

(1) Handwerk, J.H., "Ceramic Fuel Elements in the $\text{ThO}_2\text{-UO}_2$ and $\text{UO}_2\text{-PuO}_2$ Systems", Paris Fuel Elements Conf., TID-7546, Book 2, 526-47, March, 1958.

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SUMMARY OF PRELIMINARY EXPERIMENTS CONDUCTED AT ANPD

Solid solutions of UO_2 - ThO_2 were prepared in 10 w/o increments between (90 w/o UO_2 - 10 w/o ThO_2) and (10 w/o UO_2 - 90 w/o ThO_2) by reaction of UO_2 and $\text{Th}(\text{NO}_3)_4 \cdot 4\text{H}_2\text{O}$ for 4 hrs. in air at 1850°C . X-ray analysis indicated single phase FCC solid solution up to 90 w/o UO_2 . At this percentage a second phase (U_3O_8) was found to be present. The O/U ratio, as determined by lattice parameter measurements, was found to be very nearly 2.3.

Pellets of the above compositions were cold pressed at 50,000 psi and subsequently sintered in air for 2 hrs. at 1800°C . Densities varied from 85% to 90% of theoretical density for ideal ($\text{UO}_{2.3}$ - ThO_2) solid solutions. Fuel losses during this sintering process were found to range between 10 w/o and 20 w/o, assuming that all weight loss was due to loss of UO_2 .

Pellets containing 80 w/o, 30 w/o, and 10 w/o UO_2 were sintered in an argon atmosphere for 4 hrs. at 1800°C . The densities attained were 89%, 90%, and 90% theoretical density respectively.

Additions of 1 w/o MgO or 1 w/o CaO did not appear to improve the sintering characteristics of the UO_2 - ThO_2 solid solutions in inert atmospheres.

The pellets sintered in argon were subjected to a subsequent oxidation test of 24 hrs. at 2500°F in moving air. Additional weight losses of 2.3%, 0.6%, and 0.4% were observed for the pellets containing 80 w/o UO_2 , 30 w/o UO_2 , and 10 w/o UO_2 respectively. These weight losses again refer to fuel loss based on the assumption that all weight loss is due to loss of UO_2 .

Argon-sintered pellets containing 1 w/o MgO showed essentially the same weight losses as the argon-sintered ($\text{UO}_{2.3}$ - ThO_2) solid solutions when subjected to an identical oxidation treatment at 2500°F .

THERMAL SHOCK RESISTANCE OF ThO_2 AND UO_2 - ThO_2 BODIES

Thoria has very poor resistance to thermal shock and stress because of its modulus of elasticity, relatively high thermal expansivity, and low thermal conductivity. Experiments have been conducted at Armour Research Foundation⁽²⁾ in which metallic fibers, such as niobium or molybdenum, were incorporated in ThO_2 bodies. Such experiments have indicated that thermal shock resistance is improved and thermal conductivity increased in this manner. However, examination of these bodies after fabrication revealed numerous microcracks radiating from the metallic inclusions. Furthermore, upon heating in an oxidizing atmosphere for several hours at 1000°C the niobium or molybdenum fibers were found to be completely oxidized. Similar results were observed for UO_2 - ThO_2 bodies containing up to 50 w/o UO_2 .

(2) Baskin, Arenberg, and Handwerk; "Thoria Reinforced by Metal Fibers", Am. Ceramic Society Bulletin, Vol. 38, No. 7, 1959.

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CONCLUSIONS

- 1) Solid solutions of (UO₂-ThO₂) containing up to 30 w/o UO₂ show some promise at 2500°F from the standpoint of stability in air. Further evaluation of the system for higher temperature applications is necessary.
- 2) Bodies fabricated from ThO₂ or (UO₂-ThO₂) solid solution exhibit poor thermal shock resistance which severely limits their applicability as structural components in systems in which thermal cycling is to be encountered.

FUTURE EXPERIMENTS

Work is to continue in an attempt to attain pellet densities equivalent to those obtained at Argonne. These pellets will be evaluated with regard to stability in air by heating in a moving air stream at temperatures $\geq 2500^{\circ}\text{F}$. Particular emphasis will be placed on compositions containing more than 50 w/o ThO₂.

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