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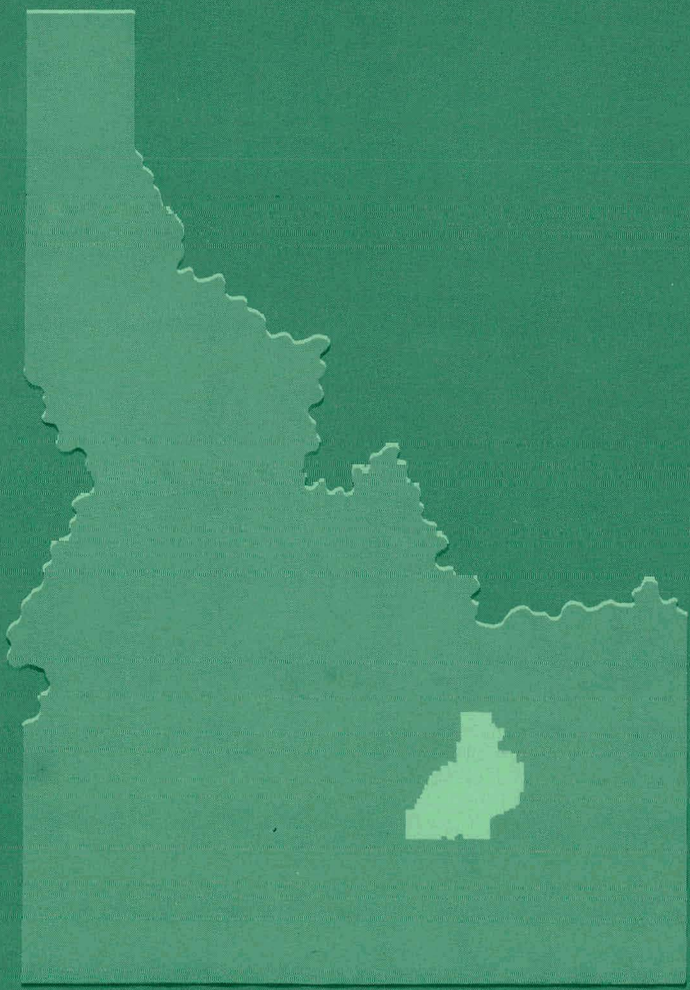

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By

J. F. Koenig

PHILLIPS PETROLEUM COMPANY
Atomic Energy Division
Idaho Falls, Idaho
Contract AT(10-1)-205

IDAHO OPERATIONS OFFICE
U. S. Atomic Energy Commission

# THERMAL STABILITY OF GADOLINIUM NITRATE SOLUTION AT HIGH TEMPERATURE


## ABSTRACT

During a feasibility study of auxiliary shutdown systems for the Spert II and Spert III reactor facilities, salts possessing high neutron absorption cross sections were considered for use in water soluble poison injection systems. One of these salts, gadolinium nitrate, appeared to be very promising. However, very little information was available in the literature on the use of gadolinium salts in stainless steel systems at high temperatures.

Tests were carried out in a high pressure static autoclave to determine the stability of gadolinium nitrate solution at elevated temperature. At temperatures of 300°F and above, the solution decomposed with formation of a white precipitate. Thus, gadolinium nitrate was found to be unacceptable for use as a soluble poison in high temperature water moderated reactor systems.


## INTRODUCTION

The Spert II and Spert III reactors are experimental facilities operated as part of the Spert program for the study of reactor safety and reactor kinetic behavior(1). Both reactors are the forced convection, pressurized-water type and utilize heterogeneous, enriched, plate-type fuel assemblies. Spert II is designed for operation at temperatures up to 400°F, and Spert III has a maximum operating temperature of 650°F. For both the Spert II and Spert III systems, reactor startup, operation, and shutdown is accomplished by eight mechanical-pneumatic fuel-poison type control rods. Investigations have recently been conducted on auxiliary means of shutting down the reactors in case the mechanical-pneumatic control rods fail while in high temperature service.

A convenient means to provide additional reactivity shutdown margin is to add a solution containing a water soluble neutron poison to the reactor coolant. The following properties would be required of the poison: high solubility in water, high neutron absorption cross section, high electrode potential and thermal stability. Gadolinium salts appeared to be desirable from the standpoint of solubility and neutron cross section. A literature survey[2,3,4,5,6] revealed that information on the thermal stability and electrode potential for gadolinium salts at elevated temperatures was generally lacking. The electrode potential of gadolinium

$$Gd \rightarrow Gd^{+3} + 3e^{-}$$

is 2.4 ev (hydrogen as zero) for 25°C and 1 molar solution[7]. Since this potential is significantly above the potentials of the elements in the stainless steel system, there should be no deposition of gadolinium on the stainless steel surface through plating action. To substantiate this assumption, and to investigate the thermal stability, an experiment was carried out using a bomb type apparatus in a static autoclave at environmental conditions simulating those of the reactor.

## SELECTION OF THE ANION

The gadolinium salts were available from the supplier in the forms of oxalate, oxide, chloride, nitrate, sulfate, and fluoride. The oxalate and oxide are relatively insoluble and, therefore, were omitted from consideration. The chloride and fluoride were omitted because of their corrosion effects on the stainless steel system[8]. The sulfate ion was eliminated since the solubility decreased with increased temperature. The nitrate salt was therefore selected for study.

## TEST DESCRIPTION

A bomb apparatus was constructed of an 8 in. length of 3/4 in. 304 stainless steel tubing with the ends sealed by compression type connectors and pipe plugs. A solution containing 10 g/l of gadolinium nitrate was prepared. This concentration, approximately 10 times that which would be required in the reactor system, was used so that any decomposition would be readily noticeable. Sufficient solution was placed in the bomb apparatus to fill it approximately half full. The charged bomb was placed in a 1 gallon static autoclave and the autoclave was brought to operating conditions. Temperature was maintained for at least 7 hours in each of the tests. At the completion of each test, the autoclave was cooled sufficiently to permit disassembly and the bomb apparatus was removed. The test solution was inspected visually for precipitation or crud. The stainless steel bomb was inspected visually to ascertain the presence of any deposition on its surface.

## RESULTS AND CONCLUSION

The results are listed in the following table:

TABLE 1

TEMPERATURE STABILITY OF GADOLINIUM NITRATE SOLUTION

| Test No. | Temp. °F | Formation of Precipitate in Solution | Formation of Film on Bomb Walls |
|---|---|---|---|
| 1 | 400 | Yes | None Observed |
| 2 | 300 | Yes | None Observed |
| 3 | 250 | No | None Observed |
| 4 | 275 | No | None Observed |

Further temperature resolution was not attempted since the autoclave controller was only capable of maintaining a constant temperature within ± 10°F.

It was found that the pH of the solution containing the precipitate was 6.4 and that the precipitate dissolved upon the addition of a small quantity of nitric acid. Therefore, an additional test was conducted with 10 g/l solution of gadolinium nitrate with the pH adjusted to 2 by the addition of nitric acid. The acidified solution also formed an insoluble precipitate upon heating. A final test was conducted with a gadolinium nitrate solution concentration of 1 g/l, the calculated concentration required to adequately poison the reactor. The gadolinium nitrate decomposed as before.

It can be concluded that gadolinium nitrate is unsuitable for use as a soluble poison in high-temperature, water-moderated reactor systems due to its thermal instability.

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