

CONF-791049--28

# Lawrence Livermore Laboratory

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SEPTEMBER, 1979

**MASTER**

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This paper was prepared for submission to:  
 23rd Conference on Analytical Chemistry in Energy Technology  
 Oak Ridge National Laboratory, Oak ridge, TN  
 October 8-10-79

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## USE OF RADIOACTIVE TRACERS FOR SELECTION OF RARE EARTH PRECIPITANTS AND IGNITION TEMPERATURES\*

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### ABSTRACT

We have found variations in the specific activity of ignited radioactive-labeled rare earth oxide samples. The variations appear to depend on the precipitating agents and temperatures. Using various precipitating agents and different ignition temperatures, samples of  $^{88}\text{Y}$ -,  $^{168}\text{Tm}$ -, and  $^{173,174}\text{Lu}$ -labeled oxides were produced from stock solutions. Observed specific activities were compared to the known specific activities of the starting solutions. At  $800^\circ\text{C}$  ignition temperatures, errors of 6% to 7% can be obtained for yttrium precipitated with 8-hydroxyquinoline or ammonia. Thulium and lutetium quinolates and cupferrates ignited at  $800^\circ\text{C}$  are in error by 3% to 5%. Our results show that temperatures in excess of  $1000^\circ\text{C}$  are required for complete ignition of rare earth quinolates, cupferrates, hydroxides, and chlorides.

### INTRODUCTION

Experiments are being done at Livermore to determine optimum conditions for precipitating and igniting some rare earth elements and yttrium to their oxides.

The possibility of conventional gravimetric rare earth assays<sup>1,2,3,4</sup> giving incorrect values was first noticed with yttrium. Lawrence Livermore Laboratory and Los Alamos Scientific Laboratory radiochemists independently became aware of the problem and came to the same conclusions as to the causes.

\*Work performed under the auspices of the U.S. Department of Energy by the Lawrence Livermore Laboratory under contract number W-7405-ENG-43.

These two laboratories use  $^{88}\text{Y}$  in their nuclear device diagnostic work. The radioisotope is mixed with a known amount of stable yttrium carrier, then various purification steps are performed to remove interfering radioactivities and mass contaminants, and finally the  $^{88}\text{Y}$ -labeled  $\text{Y}_2\text{O}_3$  is radioassayed. This type of procedure has been used for many years. Recently, however, the development of  $\text{Ce}(\text{Li})$  pulse-height-analyzers has permitted  $^{88}\text{Y}$  counting of solutions containing many other radioactivities, without the need to purify the yttrium fraction. When the  $^{88}\text{Y}$  content of purified, ignited  $^{88}\text{Y}_2\text{O}_3$  samples was compared to  $^{88}\text{Y}$  counted directly in solution with no purification, the oxide samples gave 5% to 10% lower values than direct counting.

After exploring several possible causes of the disagreement, the major source of error was found to be precipitation-ignition variations.

#### EXPERIMENTAL AND RESULTS

High-purity yttrium metal, and thulium and lutetium oxides, were used along with added radioactive tracers to prepare three stock solutions. One contained  $^{88}\text{Y}$ , another  $^{168}\text{Tm}$ , and a third solution was  $^{173,174}\text{Lu}$ , all with known amounts of tracer and carrier. The specific activity (atoms of tracer per milligram of carrier) was therefore known.

We used various precipitants and different ignition temperatures to produce  $^{88}\text{Y}_2\text{O}_3$ ,  $^{168}\text{Tm}_2\text{O}_3$ , and  $^{173,174}\text{Lu}_2\text{O}_3$  samples for counting. Table I shows the sample preparation flow chart.

TABLE I

SAMPLE PREPARATION FLOW CHART

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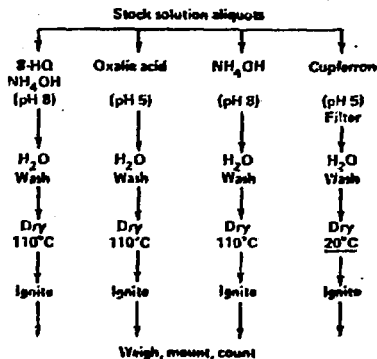


Table II shows experimental conditions and results for yttrium samples.

TABLE II

YTRIUM PRECIPITANT-IGNITION TEMPERATURE RESULTS  
(PERCENT OF THEORETICAL SPECIFIC ACTIVITY) [2]

	800°C	800°C	1000°C	1100°C	1120°C	1250°C
8-hydroxyquinoline	93.9%	98.1%	98.0%	99.8%	97.8%	—
Oxalic acid	98.7%	100.3%	99.8%	100.2%	99.8%	100.0%
NH <sub>4</sub> OH	83.2%	97.7%	98.2%	98.3%	98.2%	—
Cupferron	—	—	99.8%	—	—	99.9%

30 mins at temperature

During the time of the <sup>88</sup>Y disagreement, we were precipitating yttrium with 8-hydroxyquinoline and igniting at 800°C for 20 to 30 minutes. The results shown in Table II explain the 5% to 10% discrepancies we had noticed.

Our conclusions for yttrium are that precipitation with oxalic acid and ignition at 1000°C for 30 minutes is satisfactory.

Since other rare earths may be affected similarly, we repeated the work, using thulium and then lutetium radioactive-labeled stock solutions. Table III shows our results for thulium. No marked low results are apparent for thulium as there were with yttrium.

TABLE III

THULIUM PRECIPITANT-IGNITION TEMPERATURE RESULTS (PERCENT OF THEORETICAL SPECIFIC ACTIVITY) [2]

	800°C	1000°C	1100°C	1250°C	Fisher burner
8-hydroxyquinoline	97.3%	99.4%	99.8%	—	97.0%
Oxalic acid	98.9%	99.9%	99.3%	99.9%	—
Cupferron	98.9%	98.8%	98.7%	98.6%	97.8%

30 mins at temperature

Table IV shows lutetium results. These data show a definite temperature dependence, requiring ignition temperatures of more than 1000°C to approach a constant specific activity. Where yttrium and thulium oxalates produce analytical quality oxides at the lower temperatures, lutetium oxalate ignitions gave slightly low results from 800°C to 1250°C. This is unexpected, and will be verified as soon as possible.

TABLE IV

LUTETIUM PRECIPITANT-IGNITION TEMPERATURE  
RESULTS (PERCENT OF THEORETICAL  
SPECIFIC ACTIVITY)

	800°C	1000°C	1100°C	1250°C
8-hydroxy-quinoline	94.9%	96.6%	98.6%	98.3%
Oxalic acid	95.7%	96.0%	96.8%	96.8%
Cupferron	95.6%	96.6%	99.0%	100.3%

30 mins at temperature

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