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MEASUREMENT OF IMPURITY EFFECTS IN THE MASS SPECTROMETER ISOTOPIC ANALYSIS OF URANIUM HEXAFLUORIDE

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ABSTRACT

Tests to determine the extent of the isotopic analysis errors resulting from ion beam distortion by gaseous impurities in uranium hexafluoride samples were run on several mass spectrometers. Mixtures of uranium hexafluoride with five other gases were compared to pure uranium hexafluoride of identical isotopic composition, noting the bias of the measured ratios of mole ratios from unity.

Nitrogen caused a greater bias than the other gases tested; ten per cent nitrogen can cause a bias of more than one per cent in the ratio of pure to impure sample mole ratios.

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INTRODUCTION

The presence of gaseous impurities can affect the accuracy of the isotopic analysis of uranium hexafluoride with the mass spectrometer. This phenomenon is well known (1), but detailed information is not available. Experiments are described in this report in which the extent of the bias was determined when known levels of impurities were present.

The exact mechanism by which the effect functions is uncertain. However, when a beam of positive ions moves through a column of gas, a number of different processes of interaction between ions and gas molecules may occur: neutralization of the ions, either from the capture of a free electron or an ionization of the gas molecule; spreading of the ion beam by multiple scattering with gas molecules; or retardation of the ion beam due to collision energy loss. A space charge effect, if present, results in a shift of the position at which focus occurs. Another report (3) treats these processes in greater detail.

This study is primarily of interest in monitoring gas streams where impurities cannot be removed, as routine sample purification is practiced in the isotopic analysis laboratory. Some impurities (such as some of the fluorocarbons) are difficult to remove completely, and small inleakage of air may occur at any time, however.

PROCEDURE

Mixtures of uranium hexafluoride and five other gases, simulating impurities, were prepared by a partial pressure method, using a 0 - 100 mm. Hg Pressure Blind Multiplier (Taylor Instrument Co.) attached to the spectrometer manifold (2).

Samples of these gas mixtures were then compared with other samples having identical isotopic composition but no impurity, using routine isotopic analytical methods with the exception that sample purification steps were omitted. Thus, any apparent difference in observed isotopic content was attributed to the presence of the impurity.

Various blends of nitrogen and uranium hexafluoride were analyzed in different types and modifications of spectrometers to compare individual instrument characteristics.

All tests were made using uranium of natural of near-natural isotopic composition, except for one test in which uranium enriched in U^{235} was used.

RESULTS AND DISCUSSION

Bias

Figure 1 shows that 10 per cent nitrogen can cause a bias of more than 1 per cent in the ratio of mole ratios* of the two samples. Figure 2 displays the effects of other impurities and demonstrates that the presence of any foreign gas is undesirable.

The minimum amounts of these impurities required to raise the spectrometer ion gage reading 0.1×10^{-7} mm. Hg, and thus be detectable, range from 0.1 to 0.3 mole per cent. Quantities larger than these are usually removed before routine analyses are made, of course.

It has generally been assumed that the presence of impurities spreads the ion beams symmetrically so that the larger beam adds to the smaller one, as a net effect. Figure 2 shows that an effect opposite to this assumption occurs when measuring highly enriched uranium (93 per cent U^{235}). In this case, the large U^{235} peak does not add to the U^{238}; rather the U^{235}/U^{238} ratio is increased by the presence of impurities. This indicates that symmetrical spreading of the ion beam is not always the predominant factor. Figure 4 illustrates one possible explanation of these two conditions.

Variations Between Spectrometer Types

It has been noted that the bias effect is somewhat dependent on the exact focus condition of the spectrometer. The data in table 1 were secured with the focus used for routine analyses; this focus must frequently be a compromise between sensitivity and resolution requirements.

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* This ratio measured by the spectrometer is

$$\frac{\left(\frac{U^{235}}{1-U^{235}}\right)\ \text{Sample Plus Impurity}}{\left(\frac{U^{235}}{1-U^{235}}\right)\ \text{Pure Sample}}$$

when the U^{238} isotope is predominant. But since the dual collector-amplifier arrangement of the spectrometer requires passing the less abundant of two isotopes through the resolving slit, the ratio measured is

$$\frac{\left(\frac{U^{238}}{1-U^{238}}\right)\ \text{Sample Plus Impurity}}{\left(\frac{U^{238}}{1-U^{238}}\right)\ \text{Pure Sample}}$$

when the U^{235} isotope is the more abundant.

With the ion source of the Consolidated 21-221 spectrometer (entry 2 in table 1) immersed in the main magnetic field, it was thought that any isotope separation in the source itself might result in selective removal of ions by the second collimator slit, if its position were not perfectly centered in the beam. Consequently, this collimator was removed, and the data in entry 3 of table 1 were secured. A slight improvement is indicated from a comparison of the two tests, but further extensive measurements would be necessary to establish a significant difference of this magnitude.

The data from both 21-320A spectrometers were secured when dry ice was used for cooling the cold traps, and the analyzer pressures were about 10^{-6} mm. Hg. In all other tests liquid nitrogen was used as the coolant and base pressures were in the 10^{-7} mm. region.

The twelve-inch radius spectrometer constructed in this laboratory exhibits less bias from impurities than the other types available for the comparison, as shown in table 1. This is not surprising, in view of the greater dispersion, higher resolving power, and differential pumping system of the larger instrument.

TABLE 1

COMPARISON OF THE IMPURITY EFFECT IN DIFFERENT SPECTROMETER TYPES

Spectrometer Type	Number of Determinations	Mole Per Cent Nitrogen	Observed Ratio of Ratios
1. G. E. 60° Nier Type	13	10	1.015
	5	20	1.031
2. Consolidated 21-221	2	10	1.006
	2	20	1.013
3. Consolidated 21-221 with Lower Source Plate Removed	2	10	1.005
	2	20	1.009
4. Consolidated 21-320A (Spectrometer No. 1)	3	10	1.009
5. Consolidated 21-320A (Spectrometer No. 2)	2	10	1.003
	2	20	1.008
6. 12" Radius, 60°	8	10	1.0004
	8	20	1.0012

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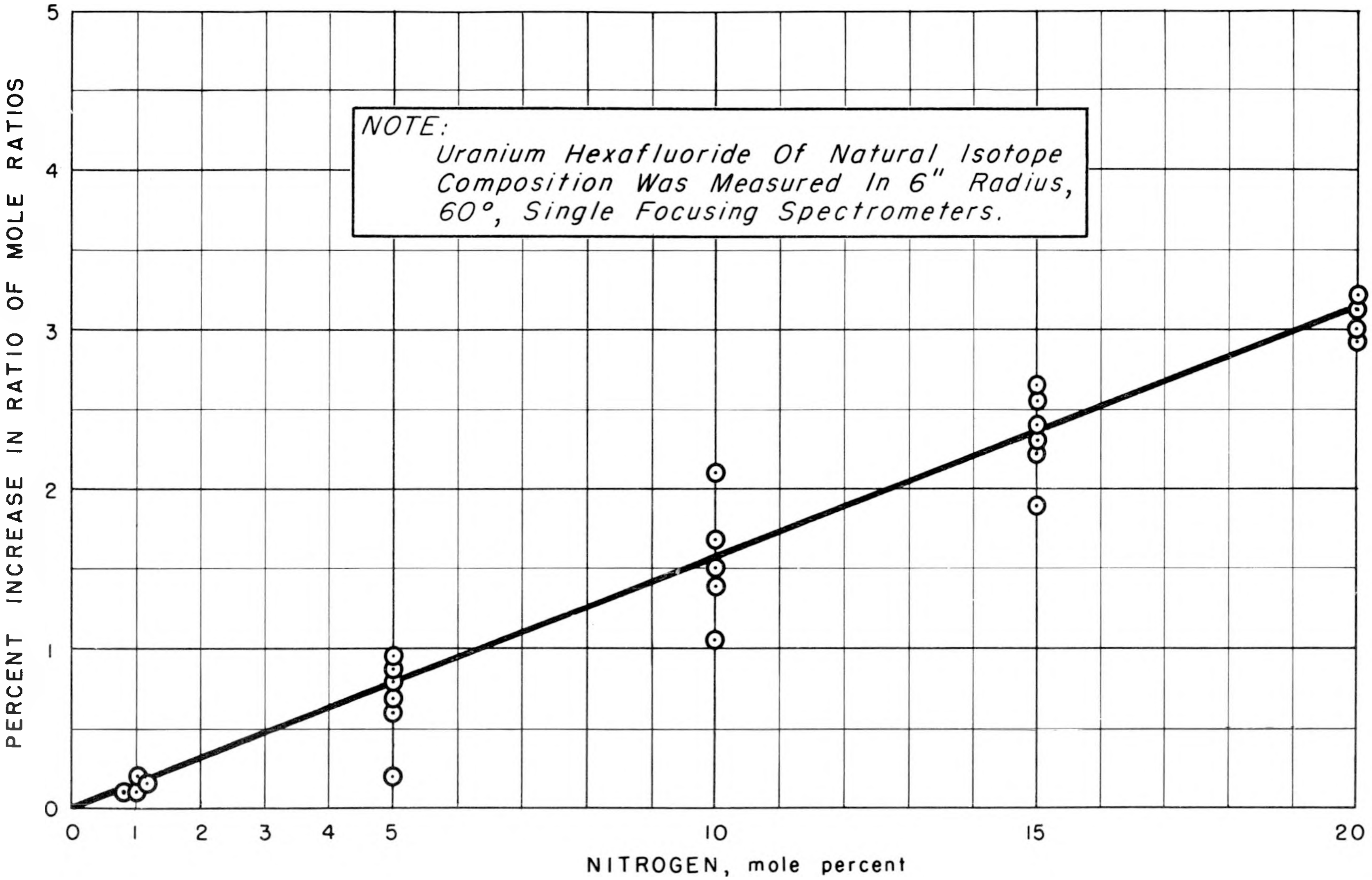
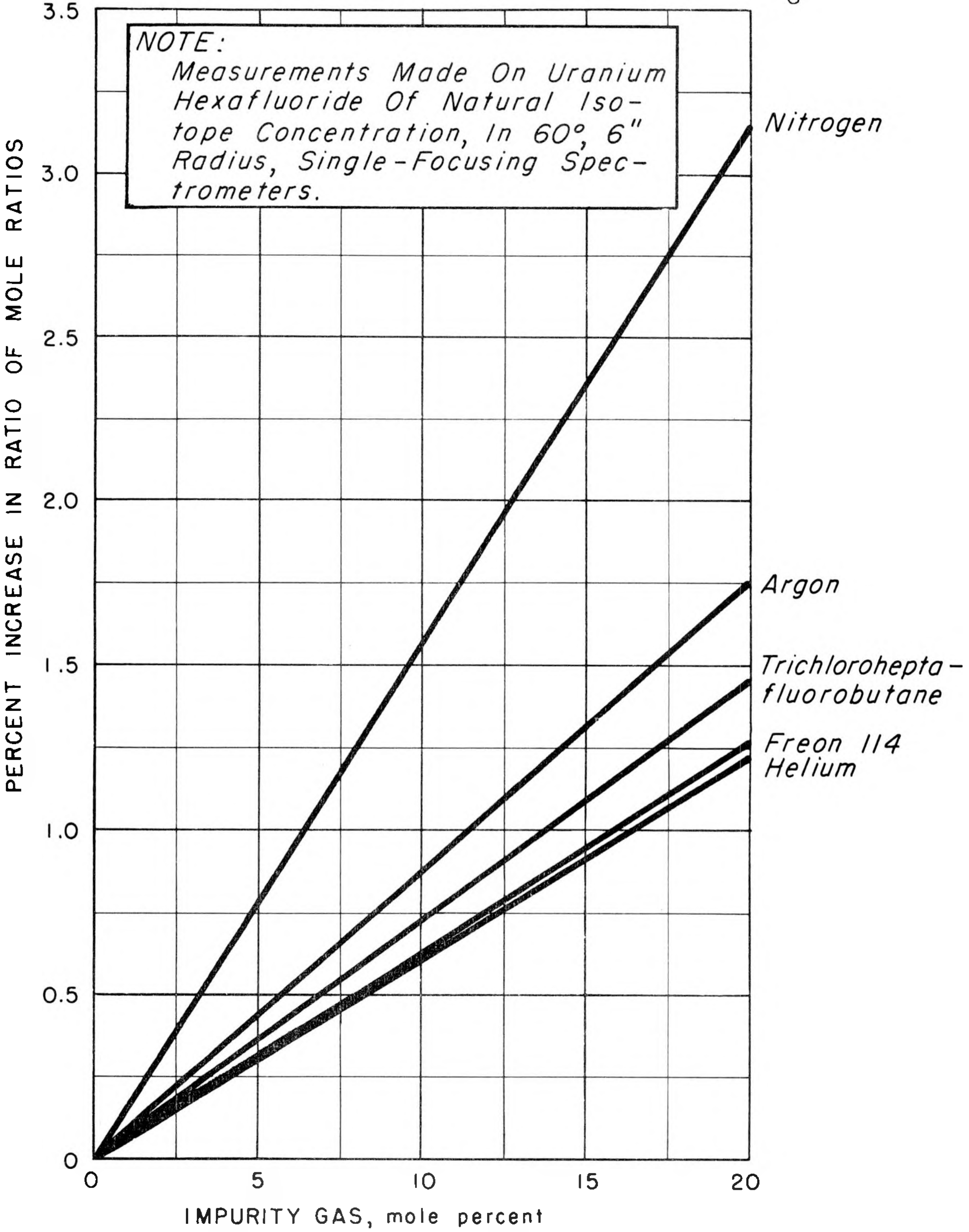


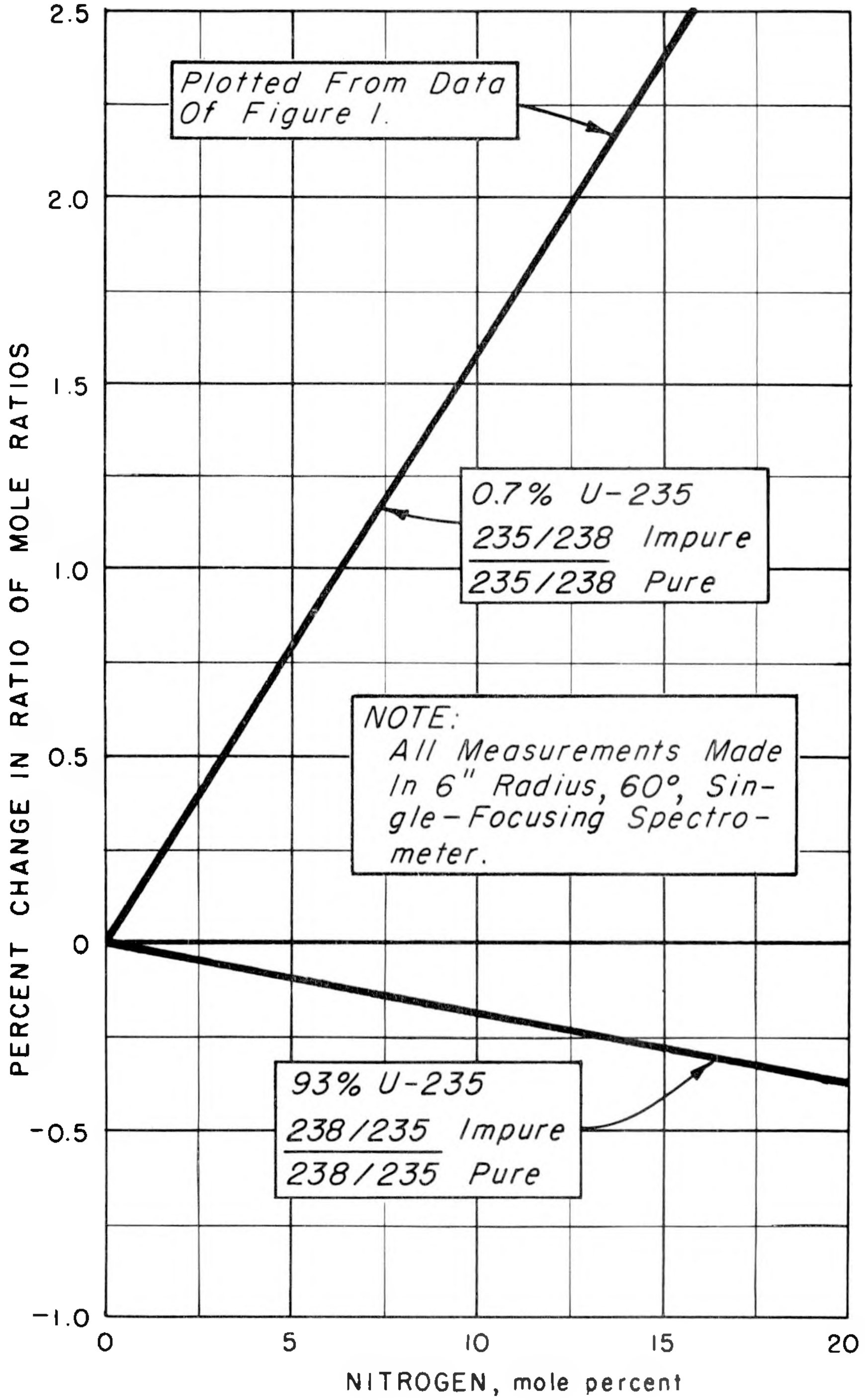
THE EFFECT OF NITROGEN ON RATIO OF RATIO MEASUREMENTS

FIGURE 1



THE EFFECT OF VARIOUS GASES ON
ISOTOPIC RATIO OF RATIO MEASUREMENTS

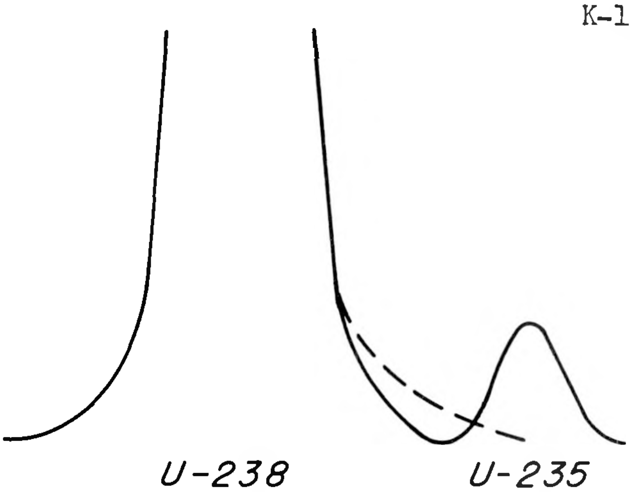
FIGURE 2



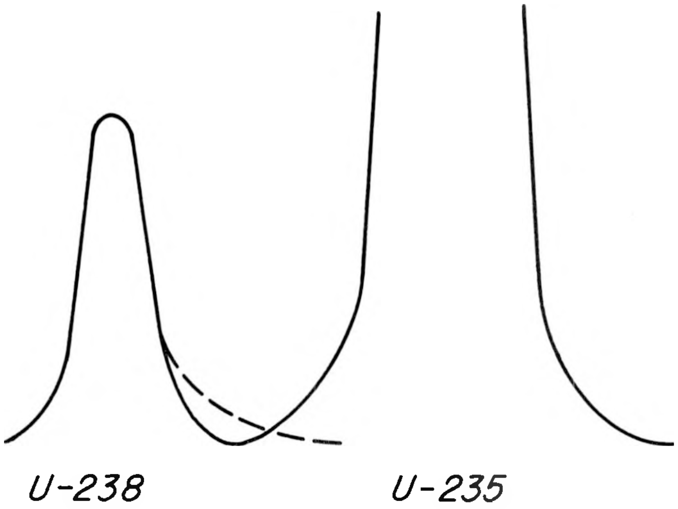
NOTE:
All Measurements Made In 6" Radius, 60°, Single-Focusing Spectrometer.

THE EFFECT OF NITROGEN ON ISOTOPIC RATIO OF RATIO MEASUREMENTS OF NATURAL VS ENRICHED URANIUM HEXAFLUORIDE

FIGURE 3



Dotted Line Shows Probable Condition When Impurities Are Present In The Natural Uranium. The Toe Of The U-238 Peak Contributes To The U-235 Peak.



Dotted Line Shows Possible Condition When Impurities Are Present In Enriched Uranium. The Toe Of The U-238 Peak Contributes To The U-235 Peak.

THE EFFECT OF IMPURITIES ON PEAK SHAPE

FIGURE 4