

Vapour pressure differences of the Xenon Isotopes

Amal Alamre*†

Carleton University

Ottawa, Canada

E-mail: AmalAlamre@cmail.carleton.ca

Another Author

A. Alamre1, I. Badhrees1,2, B. Death1, C. Licciardi3, D. Sinclair1,4

1Department of Physics, Carleton University, Ottawa Canada. 2KACST. 3Laurentian University, Sudbury, Canada. 4TRIUMF Laboratory, Vancouver Canada.

Abstract:

A central topic in the development of understanding of the nature of neutrinos is the search for neutrino-less double beta decay. As increasingly sensitive detectors are required for such searches, a major cost and feasibility concern is the availability of sufficient separated isotope for the chosen target. ^{136}Xe is a very attractive target for these studies. At present this isotope is only available from the Centrifuge systems in Russia. We are exploring the feasibility of doing this enrichment using distillation. To start this study we require the vapour pressure differences of the xenon isotopes which have not been measured to date. This paper will describe the measurement of these vapour pressures using a tall cryogenic distillation process.

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*Speaker.

†A footnote may follow.

1. Introduction

Next generation searches for neutrinoless double beta decay require large quantities of target material. In general, economic or background issues dictate use of isotopic enrichment. ^{136}Xe is a very attractive target. To reach the normal hierarchy for neutrino masses, 50 tonnes of this isotope are required. Currently the only enrichment technique available is centrifuge, and this is only possible in Russia. We are exploring the use of distillation for xenon enrichment. In this work we report the first credible measurement of the separability of xenon isotopes by distillation and hence the first measure of the xenon vapour pressure isotope effect (VPIE). A recent review of the status of VPIE measurement and theory has been published by [1].

2. The Measurement

A 1.8m high cryogenic still has been built at Carleton. It uses a 25 mm ID, vacuum insulated reflux column filled with PROPAK2) packing material. A condenser is placed at the top of the column and a re-boiler at the bottom. The still is operated at 100% reflux. All measurements were made at a pressure of 105 Pa. The vapour is sampled at the top and bottom of the packed bed using a large bore quadrupole mass spectrometer to determine the isotopic abundances. The ion detector in the spectrometer is operated in current mode rather than pulse counting mode as this reduces the non-linearity in the response. The spectrometer is sequenced to sample first the top and then the bottom and then repeat, acquiring each mass sample over a period of approximately 20 minutes. This process is continued for several days. Asymptotic values are reached after about 6 hours, and after this the continued sampling serves to improve the precision of the measurement. This process measures directly the separability of the isotopes for a given packing and operating condition. In order to produce a measure of the VPIE, one needs to calibrate the still. This is done by running first using argon and then krypton (for which measurements of the VPIE have been published) and then taking data for xenon.

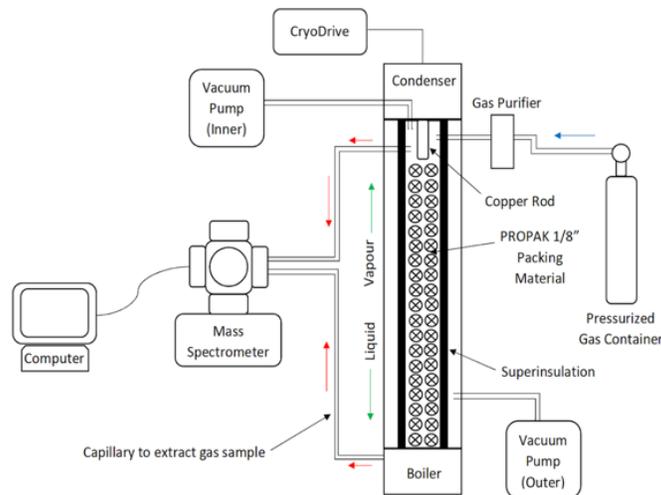


Figure 1: Experimental setup showing gas entering the top of the still, with samples extracted from the top bottom to be sent to the mass spectrum.

3. Data analysis

Typical mass spectra for the three elements are shown in Figure 1. It can be seen that each of the isotopes is cleanly separated. Shapes of the peaks are not Gaussian, so to determine the relative peak intensities, the strongest clean line is used as a template and the spectrum is then fitted as a sum of the peaks plus a constant background. Peak intensities are then normalized to the total intensity. This method of taking ratios of ratios eliminates most of the systematic errors. Figure 2 displays the resulting separation power of our still for each isotope.

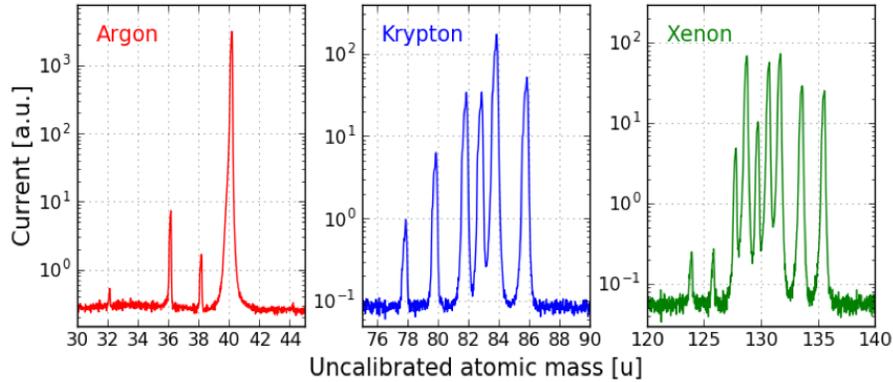


Figure 2: Mass spectra for argon, krypton and xenon.

In order to extract the VPIE values one must correct for the still performance. As a first approximation one can simply assume the height of a theoretical plate (HETP) is the same for each element. Then the VPIE's should be proportional to the slopes of the curves in Figure 2. This approximation gives reasonable agreement with previous data when comparing argon and krypton. Two corrections should be made, however. The mass transfer coefficients should vary with the square root of the vapour phase diffusion coefficients³). We calculate these from simple kinetic theory. VPIE's are normally quoted at the triple point while our measurements are made at the boiling point. We use the theoretical dependence of the VPIE on temperature ($VPIE \propto 1/T^2$) where T^* is the depth of the Lenard-Jones potential for the element⁴) to correct our data back to the triple point. The final results are presented in Table 1 together with the theoretical VPIE's.

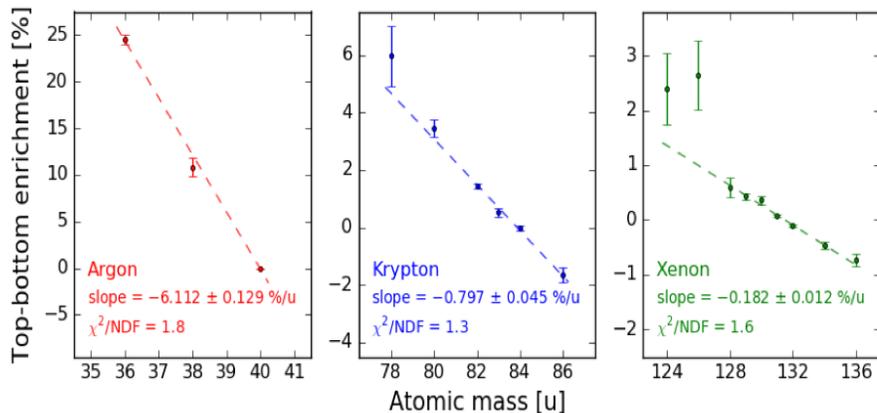


Figure 3: Fractional difference between the top and bottom of the column for each isotope.

The slope of the line determines the separation per mass unit for the still and this is proportional to the VPIE per mass unit for each element. The linear relationship with mass is expected from theory but has not been previously reported.

Element	Slop[%u]]	Mass Transfer	BP to Triple Point	Exp.VPIE at TP	Tew
Ar	-6.112	1.0	1.35	1.65E-03	1.65E-03
Kr	-0.797	1.28	1.06	2.17E-04	2.50E-04
Xe	-0.182	1.39	1.05	5.28E-05	5.17E-05

Table 1: Measured enrichment factors for the cryogenic still together with the inferred VPIE.

In Table 1, The slope column gives the slope of the enrichment factor expressed as %AMU from Figure 3. Vapour Pressure Isotope Effect (VPIE) projected to the triple point are compared with the expectation from the review by Tew¹). For this review the values for argon and krypton were experimental values while the xenon value is taken from theory. All VPIE values are changes per unit mass. The value for Ar has been used to normalize our values for Krypton and xenon. The agreement between our value for krypton compared with previous work is a measure of the accuracy of analysis.

4. Discussion

We have produced the first credible measure of the separation power of a still for xenon isotopes. These lead to the first VPIE data for xenon and these are in good agreement with the theoretical predictions. We also have demonstrated that the predicted linear relationship between VPIE and isotope mass is followed for argon, krypton and xenon. This is the first time this aspect of the theory has been tested. The results will allow a study of the feasibility of distillation as a technique for isotopic enrichment of xenon. The results are also relevant to the use of the triple point of xenon as a temperature standard.

5. Acknowledgements

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