

## CONCLUSION

In view of the large statistical errors, the author feels that the results presented here indicate, but do not prove, that double beta-decay may occur in  $\text{Zr}^{96}$  without the emission of neutrinos. Further experimentation is clearly necessary.

The author wishes to thank Professor H. W. Fulbright for suggesting this experiment and for his advice and encouragement both before and during the performance of the research. He also wishes to thank Mr. A. Petschek for many helpful discussions regarding the theoretical background of the problem.

## Argon 38 in Pitchblende Minerals and Nuclear Processes in Nature

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The isotopic constitution of argon extracted from four pitchblende samples of different age and uranium concentration has been determined. The  $A^{36}/A^{38}$  ratio was found to vary by over three hundred percent with the greatest deviations from normal occurring in the ores with high uranium content. It is suggested that  $A^{38}$  has been produced in considerable quantities in these ores by nuclear reactions involving  $\alpha$ -particles and/or the spontaneous fission neutrons.

THE occurrence of large variations in the abundance of  $A^{40}$ , because of  $K$  capture in  $\text{K}^{40}$ , in argon samples extracted from rocks is well known and has been studied in many laboratories for the purpose of age determinations. For example, Mousuf<sup>1</sup> has found values of the  $A^{40}/A^{36}$  ratio as high as 90 000 in samples of microcline while the value of this ratio for normal argon, as determined by Nier,<sup>2</sup> is 295.5. Since no detailed study of the  $A^{38}$  content of argon samples from various sources has been reported, a survey of the  $A^{38}$  abundance in pitchblende ores was undertaken in this laboratory in conjunction with the investigation of the xenon and krypton spontaneous fission yield patterns already in progress.<sup>3</sup>

Powdered pitchblende samples of approximately 200 grams were placed in an inconel combustion tube and continuously evacuated for 24 hours and then heated to 250–300°C for one hour to remove most adsorbed gases. Preliminary experiments showed that no loss of the fission product gases occurred during this period. The temperature was then slowly raised to the maximum obtainable with the furnace used (approximately 1250°C) and the gases released were collected in two or three fractions over the temperature range covered. Fractions collected at the higher temperatures should contain less normal argon from adsorbed gases. Each fraction was purified in a calcium furnace to constant volume. The volume of the individual samples varied from one to ten cubic millimeters depending on the ore used and the temperature range covered.

The samples were analyzed on a 180° direction focusing mass spectrometer. The small size of the

samples made it necessary to operate the ion source with very high ionizing electron beam currents.

The principal sources of error in determining the abundance of a rare isotope, such as  $A^{38}$ , are impurities in the sample and residual or background ion currents in the mass spectrometer. To eliminate the first factor one sample was removed from the mass spectrometer and repurified in a calcium furnace. The analyses before and after repurification were in good agreement (Table I). Increasing the peak heights by a factor of three by changing the sample pressure produced no detectable change in the abundance ratios, indicating that the effect of the background ion currents was negligible.

The results of the mass spectrometer analysis are given in Table I together with the ratios for normal argon. The uranium concentration and geological ages determined by lead isotope ratio measurements are also listed. The Great Bear Lake pitchblende result is from preliminary work and only the highest temperature fraction was available for analysis. The  $A^{36}/A^{38}$  ratio is seen to vary from 1.69 to 5.18 for the pitchblende samples, compared with 5.35 for normal argon. This is an over-all variation of more than 300 percent. Such large changes in isotopic abundance cannot be explained by any natural fractionation process and can be produced only by some nuclear reactions. The xenon and krypton fission yield patterns were found to be independent of the temperature at which the samples were collected, indicating that no appreciable fractionation results from the diffusion of the gases out of the crystal lattice during the extraction process.

It is, therefore, necessary to consider possible nuclear reactions which would result in the production of  $A^{38}$  or the removal of  $A^{36}$ . Since no correlation between the

<sup>1</sup> A. K. Mousuf, *Phys. Rev.* **88**, 150 (1952).

<sup>2</sup> A. O. Nier, *Phys. Rev.* **77**, 789 (1950).

<sup>3</sup> J. Macnamara and H. G. Thode, *Phys. Rev.* **80**, 471 (1950).

TABLE I. Argon isotope abundance ratios.

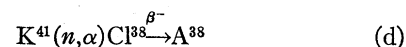
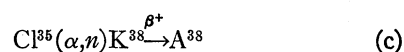
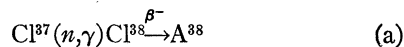
Sample	Age (millions of years)	Percent U <sub>3</sub> O <sub>8</sub>	A <sup>40</sup> /A <sup>36</sup>	A <sup>36</sup> /A <sup>38</sup>
Normal argon, Nier <sup>a</sup>	—	—	295.5	5.35
Pitchblendes				
Lake Athabaska	1686	13.99		
(1) 500–850°C			280.0±0.6 <sup>b</sup>	5.18 ±0.02
(2) 850–1050°C			291.6±1.8	4.97 ±0.06
(3) 1050–1240°C			289.0±1.0	5.14 ±0.01
Great Bear Lake				
(1) 1000–1200°C	1370	36.46 <sup>c</sup>	270.2±0.5	5.15 ±0.04
Eagle Mine, Beaverlodge				
(1) 250–900°C	1627	45.5	265.6±0.2	2.598±0.015
(1) repurified			268.2±0.6	2.582±0.012
(2) 900–1170°C			226.4±0.4	1.69 ±0.01
Belgian Congo	642	65.21		
(1) 300–880°C			294.0±1.2	3.87 ±0.04
(2) 880–1240°C			376.8±8.5	4.14 ±0.10

<sup>a</sup> See reference 2.

<sup>b</sup> Errors quoted are probable errors of the mean of the individual mass spectrometer determinations. All results are relative to Nier's values for normal argon.

<sup>c</sup> A mill concentrate, original concentration unknown.

age of the samples and the A<sup>36</sup>/A<sup>38</sup> ratio is apparent in Table I, and since no nuclides decaying to A<sup>36</sup> or A<sup>38</sup> have half-lives long enough to be of significance on a geological scale of time, the interactions of alpha-particles and the spontaneous fission neutrons with the elements in the neighborhood of argon must be considered. The amount of A<sup>38</sup> present in the samples was approximately the same as the amount of fission product Xe<sup>132</sup>. The very low relative probability of multiple fission indicates that this process cannot account for the abnormal amount of A<sup>38</sup> present. The large deviations from normal in the A<sup>36</sup>/A<sup>38</sup> ratio are found in the ores of high uranium content, where reactions involving alpha-particles and neutrons would be expected to be more prominent. The principal reactions producing A<sup>38</sup> or using up A<sup>36</sup> are listed below.



Reaction (a) may be disregarded because the capture cross section of Cl<sup>35</sup> is much larger than that of Cl<sup>37</sup>; hence more A<sup>36</sup> than A<sup>38</sup> would be produced if neutron capture in chlorine were important. Any reaction which used up A<sup>36</sup> would be accompanied by a similar reaction involving A<sup>38</sup>, probably to about the same extent, and hence reactions (e), (f), and (g) would not appear to explain the observed A<sup>36</sup>/A<sup>38</sup> ratios. Of the remaining reactions, (b) and (c) appear to be the most probable, but the data available are by no means sufficient to establish this definitely. It should be noted that reactions similar to (b) and (c), involving S<sup>33</sup>, would produce A<sup>36</sup>, and this must be taken into account. The extent to which reactions of these types occur would depend on the uranium content, on the concentration of the light elements involved, and on the age of the ore.

Further studies of argon samples from pitchblende and other rocks are in progress, and a study of the chemical composition of the pitchblende samples is under way. It is hoped that it will be possible to identify the reaction or reactions producing the abnormal A<sup>36</sup>/A<sup>38</sup> ratios.

The results obtained to date show that nuclear reactions produce quite large variations in the abundance of A<sup>36</sup> and A<sup>38</sup> in nature. This indicates that the corrections for normal argon, based on A<sup>36</sup> abundance, used in age determinations involving K<sup>40</sup> decay may be subject to large errors, particularly if uranium is associated with the materials being studied.

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