

²J. deBoer and J. Eichler, in Advances in Nuclear Physics, edited by M. Baranger and E. Vogt (Plenum Press, Inc., New York, 1968), Vol. 1.

³K. Kumar and M. Baranger, Phys. Rev. Letters **17**, 1146 (1966).

⁴For a discussion of the underlying theory see Ref. 2.

⁵J. E. Glenn and J. X. Saladin, Phys. Rev. Letters

19, 33 (1967).

⁶Aage Winther and Jorrit deBoer, in Coulomb Excitation, edited by K. Adler and A. Winther (Academic Press, New York, 1966).

⁷F. K. McGowan and P. H. Stelson, Phys. Rev. **122**, 1274 (1961), and private communication.

⁸A. Winther, private communication.

EXPERIMENTAL EVIDENCE FOR THE DOUBLE-BETA DECAY OF Te^{130} †

T. Kirsten and O. A. Schaeffer

Department of Earth and Space Sciences, State University of New York, Stony Brook, New York

and

E. Norton and R. W. Stoenner

Department of Chemistry, Brookhaven National Laboratory, Upton, New York

(Received 1 April 1968)

It is shown that double-beta decay occurs in nature. The half life of Te^{130} is $10^{21.34 \pm 0.12}$ yr.

Attempts to detect double-beta decay directly by means of coincidence techniques or nuclear emulsions have not led to an actual observation of double-beta decay.¹ Lower limits for the half lives of different isotopes were established. The most studies were made on Ca^{48} .¹

Another approach is to detect the accumulation of the decay product during geological time periods ($\sim 10^9$ yr) in those minerals which are rich in a suspected $\beta\beta$ -active isotope. Two very suitable cases for this method are the decays of $\text{Te}^{130} \xrightarrow{\beta\beta} \text{Xe}^{130}$ and $\text{Se}^{82} \xrightarrow{\beta\beta} \text{Kr}^{82}$.²

An anomalous isotopic composition of xenon extracted from tellurium minerals has been reported repeatedly.^{3,4} In all cases, surplus amounts of Xe^{129} , Xe^{130} , and Xe^{131} were superimposed on the general pattern of xenon of atmospheric composition. These findings have not been accepted¹ as proof for the double-beta decay of Te^{130} since the Xe^{130} anomaly was always accompanied by Xe^{129} and Xe^{131} excesses not yet completely understood. Rather, it was suspected that all three anomalies might result from the same unknown mechanism. In some cases, the xenon spectrum was even more confused by an additional fission xenon component which complicated the computation of the atmospheric Xe^{130} correction. The Xe^{130} excess was arrived at only by a rather involved calculation which included large uncertainties. The magnitude of the Xe^{130} excess has been less than 6% of the total xenon amount in all samples studied by other authors. In addition, the calculated half lives of Te^{130} were based on assumed geological ages of the miner-

als rather than on age determinations of the tellurium ores themselves.

To prove that Te^{130} is $\beta\beta$ active one has to show that the Xe^{130} excess is unambiguously due to double-beta decay. In addition, a calculated half-life can only be accepted if it is based on radioactive dating of the mineral.

We have analyzed the isotopic composition of xenon extracted from a native tellurium ore from the Good Hope mine (Colorado).⁵ We found a large excess of Xe^{130} not accompanied by any other anomalies.⁶ In uncrushed samples not exposed to air contamination, the excess Xe^{130} amounts to about 70% of the total xenon and the ratio $\text{Xe}_{\text{excess}}^{130}/\text{Xe}_{\text{atmospheric}}^{130}$ is higher than 50. The Xe^{130} excess does not constitute a small anomaly on the border of detectability but determines the pattern of the xenon spectrum (Fig. 1).

The absence of other xenon anomalies rules out processes other than double-beta decay which might possibly result in a Xe^{130} excess.^{7,2} The production of Xe^{130} via two successive single-beta decays is impossible since the mass excess of I^{130} is 477 ± 35 keV larger than the mass excess of Te^{130} .⁸ We must then conclude that the Xe^{130} excess is due to double-beta decay of Te^{130} .

From the amount of $\text{Xe}_{\text{excess}}^{130}$, the tellurium concentration, and the gas-retention age of the mineral, the half-life of Te^{130} can be calculated. We determined the K-Ar age of the ore and obtained an age of 1.31 ± 0.14 Gyr. The age is considered to be reliable since it is consistent with the geological situation of the ore deposit.⁹

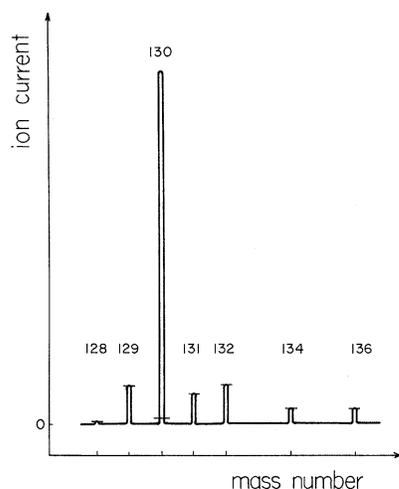


FIG. 1. Isotopic composition of xenon extracted from native tellurium ore (run No.2). The horizontal lines indicate the maximum contribution of atmospheric xenon.

The resulting half-life of Te^{130} is $10^{21.34 \pm 0.12}$ yr and is in agreement with the theoretically predicted¹⁰ half-life for the lepton-conserving two-neutrino mode of decay ($T_{1/2}^{130}, 2\nu, \text{theor} = 10^{22.5 \pm 2.5}$ yr). However, mainly because of the uncertainty in the theoretical prediction, a slight contribution of the lepton-nonconserving neutrinoless

decay mode (predicted half-life $T_{1/2}^{130}, \text{no } \nu, \text{theor} = 10^{16.3 \pm 2.2}$ yr)¹⁰ cannot be ruled out. The maximum interaction amplitude which does not conserve leptons is limited by $\alpha \leq (T_{1/2}^{130}, \text{no } \nu, \text{theor} / T_{1/2}^{130}, \text{exp})^{1/2} \approx 10^{0.5(16.3-21.3)} \approx 3 \times 10^{-3}$.

Experimental details.—The available sample was a compact piece of fresh-appearing native tellurium, 17 g in weight. It was supplied to us by Professor C. Frondel from the Harvard mineral collection (sample No. 98589). The surface was cleaned with a dental drill. The sample was then broken and two solid inner pieces (about 1 g each) were used for the mass spectrometer runs Nos. 1 and 2. The remaining 15 g were crushed and screened to yield 12 g of ore having grain sizes between 62 and 405 μ . This sample was split in two parts. One part was used for the chemical analysis. From the other part, three samples were prepared for the mass spectrometer runs Nos. 3, 4, and 5. The chemical bulk analysis gave $99.4 \pm 0.6\%$ Te with a trace of Fe.¹¹ K and U¹² as well as Th were determined by neutron activation. The results were 5.6 ± 0.2 ppm K, 21.0 ± 2 parts per billion (ppb) U, and ≤ 10 ppb.

The techniques involved in handling and analyzing minute amounts of rare gases by mass spec-

Table I. Results of the rare-gas analysis.

Run	Sample	Extraction Time (min)	Temp. (°C)	Rare Gas Amounts (10^{-8} cc STP/gram)						$\frac{\text{Xe}_{\text{rad}}^{130}}{\text{Xe}_{\text{atm}}^{130}}$
				Radiogenic Gases		Gases of Atm. Composition ^a				
				He ⁴	Ar ⁴⁰	Xe ¹³⁰	Ar	Kr	Xe	
1	0.880 g whole piece	10	1100	10.3	1.03	0.00283	4.0	0.00049	0.00141	48.1
2	0.866 g whole piece	10	1100	40.4	1.56	0.00272	23.9	0.00095	0.00118	56.5
3 a	0.894 g 62 μ -405 μ	~ 1	$\sim 600^b$	42	4.27	0.00233	36.1	0.01340	0.01690	3.38
b		10	1100	2.7	0.37	0	2.1	0.00123	0.00248	—
total				44.7	4.64	0.00233	38.2	0.01463	0.01938	2.95
4 a	1.745 g 62 μ -405 μ	800	200	2.1	0	0.00011	15.9	0.00554	0.00298	0.90
b		800	200	2.5	0.15	0.00002	4.1	0.00168	0.00136	0.36
c		270	310	12.0	1.02	0.00082	8.7	0.00503	0.00593	3.39
d		10	1100	34.1	2.89	0.00118	10.7	0.00368	0.00255	11.4
total				50.7	4.06	0.00213	39.4	0.01593	0.01282	4.07
5 a	0.779 g 62 μ -405 μ	1400	200	3.0	0	0.00015	16.4	0.00575	0.00422	0.87
b		10	1100	52.0	4.12	0.00184	22.1	0.01025	0.00737	6.11
total				55.0	4.12	0.00199	38.5	0.01600	0.01159	4.21
3-5	weighted averages			50.3	4.22					
				± 6	± 0.4					
1-5	weighted average					0.00240				
						± 0.0004				

^aExcept for He⁴, Ar⁴⁰ and Xe¹³⁰ no other isotopic anomalies have been found.

^bNative tellurium melts at 452°C.

trometry are described, except for one improvement,¹³ elsewhere.^{14,2,4} In runs Nos. 1 and 2, we extracted the rare gases in only one step by melting the ore. In runs Nos. 3, 4, and 5 the gases were extracted stepwise as described in Table I which contains the results. The mean value for Xe¹³⁰ excess is $(2.4 \pm 0.4) \times 10^{-11}$ cc STP/g; the radiogenic Ar⁴⁰ content of the crushed sample is $(4.22 \pm 0.4) \times 10^{-8}$ cc STP/g, and the He⁴ content is $(50.3 \pm 6) \times 10^{-8}$ cc STP/g.

Runs Nos. 1 and 2 had the lowest yield of atmospheric xenon since no inner surfaces were exposed to air contamination. In these cases, the Xe¹³⁰ anomaly was about 5000%. For dating purposes it is absolutely necessary to prepare a finely-crushed and well-mixed average sample in order to exclude the effects of K and U inhomogeneities. This necessity is underlined by the different radiogenic Ar⁴⁰ contents of the crushed and the uncrushed samples. The amount of the atmospheric xenon due to surface adsorption is higher in the crushed samples but becomes relatively lower after outgassing at moderate temperatures (Table I). The absolute amounts of excess Xe¹³⁰ are in reasonable agreement for all five runs. From the data we obtained the following gas-retention ages: K-Ar age = 1.31 ± 0.14 Gyr; U-He⁴ age = 205 ± 55 Myr. The results indicate that the thermal history of the ore was such that helium is partially lost by diffusion but not Ar. If this is true, one may then conclude that probably no Xe¹³⁰ loss occurred. A high Xe¹³⁰ retentivity is also supported by the stepwise heating experiments. The half-life for the double-beta decay of Te¹³⁰ is then $10^{21.34 \pm 0.12}$ yr.

Despite the fact that $\beta\beta$ decay was not observed with certainty, half-lives for Te¹³⁰ have been calculated previously.³ Based on geological assumptions for the gas-retention ages of the tellurium ores, half-lives between $10^{20.48}$ yr and $10^{21.15}$ yr have been reported.³ The true ages of these minerals can now be calculated by introducing the Te¹³⁰-Xe¹³⁰ age-determination method, based on the half-life obtained in this work.

We wish to thank Professor C. Frondel for providing us with the rare mineral, Dr. Z. Peterman, Dr. J. Olson, Dr. D. Hedlund, and Professor S. Goldich for valuable information concerning the geology of the ore deposit, and Dr. R. Davis for helpful suggestions.

†Work supported by the U. S. Atomic Energy Commission, partially under Contract No. At-(30)-3629.

¹Earlier work reviewed by S. P. Rosen and H. Prima-

koff, in Alpha-, Beta-, and Gamma-Ray Spectroscopy, edited by K. Siegbahn (North-Holland Publishing Company, Amsterdam, The Netherlands, 1965), Vol. 2, p. 1499; and by V. R. Lazarenko, *Usp. Fiz. Nauk* **90**, 601 (1966) [translation: *Soviet Phys. Usp.* **9**, 860 (1967)]. For recent references see R. K. Bardin, P. J. Gollon, J. D. Ullman, and C. S. Wu, *Phys. Letters* **26B**, 112 (1967); C. Y. Chang, G. B. Yodh, R. Ehrlich, R. Plano, and A. Zinchenko, *Phys. Rev. Letters* **20**, 510 (1968).

²T. Kirsten, W. Gentner, and O. A. Schaeffer, *Z. Physik* **202**, 273 (1967).

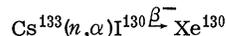
³M. G. Inghram and J. H. Reynolds, *Phys. Rev.* **76**, 1265 (1949), and **78**, 822 (1950); N. Takaoka and K. Ogata, *Z. Naturforsch.* **21a**, 84 (1966); E. K. Gerling, J. A. Shukoljukov, and G. S. Askinadze, *Yadern. Fiz.* **6**, 311 (1967) [translation: *Soviet J. Nucl. Phys.* **6**, 226 (1968)], and *Geokhimiya* **9**, 1038 (1967).

⁴T. Kirsten, W. Gentner, and O. Müller, *Z. Naturforsch.* **22a**, 1783 (1967).

⁵A part of the data of this investigation has been published in a preliminary state (Ref. 2). Reference 2 contains a detailed discussion of the principles, methods, and techniques of mass spectrometric detection of $\beta\beta$ -decay products and of their theoretical interpretation.

⁶The results show that the Xe¹²⁹ and Xe¹³¹ excesses observed in other tellurium ores are due to the contamination of the samples by U. Measured U concentrations in other tellurides are 196 and 225 ppm (Refs. 3 and 4). This is four orders of magnitude above the U concentration in the sample discussed here. A further discussion of the Xe¹²⁹ and Xe¹³¹ anomalies in U-rich tellurides may be found elsewhere.^{2,4}

⁷If Xe¹³⁰ were produced by spontaneous fission of uranium, one should see about 10^5 times more Xe¹³⁶ than Xe¹³⁰. Cosmic-ray-induced spallation reactions would produce much more Xe¹²⁴ and Xe¹²⁶ than Xe¹³⁰, but no Xe¹²⁴ and Xe¹²⁶ anomalies are observed. In addition, the ore was shielded by the atmosphere and rocks above the mine. Meson-induced reactions do not lead to Xe¹³⁰. Neutron-induced reactions would primarily form Xe¹²⁹ and Xe¹³¹, but no such anomalies exist in our sample. The absence of Xe¹²⁹ and Xe¹³¹ anomalies sets limits on the maximum internal neutron flux. From this limit and the highest possible estimate of the cesium concentration, it follows that the maximum contribution of the reaction



is far below the detection limit. Natural α radiation would produce Xe¹²⁸, Xe¹²⁹, and Xe¹³¹ but not Xe¹³⁰, since natural α energies are insufficient for the reaction $\text{Te}^{128}(\alpha, 2n)\text{Xe}^{130}$.

⁸ $\Delta M(\text{I}^{130}) = -868.90 \pm 19$ keV, H. Daniel, M. Kuntze, B. Martin, P. Schmidlin, and H. Schmitt, *Nucl. Phys.* **63**, 145 (1965). $\Delta M(\text{Te}^{130}) = -873.67 \pm 16$ keV, N. Zeldes, M. Gronau, and A. Lev, *Nucl. Phys.* **63**, 1 (1965).

⁹The Good Hope Mine (Vulcan mining district, N38°20' 45" - 107°00' 15" W) belongs geologically to the Gunnison River (Colorado) Series. Veins rich in pyrite, gold and copper tellurides, and native tellurium

occur in Precambrian amphibole schists (Dubois greenstone); E. B. Eckel, U. S. Geol. Surv. Bull. 1114, 1 (1961); J. F. Hunter, U. S. Geol. Surv. Bull. 777, 1 (1925). No radioactive dating on the Precambrian basement near Vulcan has been done. However, for the basement in the nearby Monarch Pass-Gunnison area, Rb-Sr ages between 1.3 and 1.6 Gyr have been reported by G. W. Wetherill and M. E. Bickford, *Trans. Am. Geophys. Union* 45, 116 (1964). The stage of mineralization in the Vulcan district is probably also Precambrian [W. S. Burbank and C. T. Pierson, U. S. Geol. Surv. Circ. 236, 1 (1953); Z. Peterman, J. Olson, and D. Hedlund, U. S. Geological Survey, Denver, Colorado, private communication].

¹⁰Calculated from the formula given by Rosen and

Primakoff (Ref. 1) and new mass excess data by N. Zeldes, M. Gronau, and A. Lev, *Nucl. Phys.* 63, 1 (1965). The mass difference between Te^{130} and Xe^{130} is 2509 ± 19 keV.

¹¹A similar result on a native tellurium sample from the Good Hope Mine is reported by W. P. Headden, *Colo. Sci. Soc. Proc.* 7, 141 (1903).

¹²R. W. Stoenner and J. Zähringer, *Geochim. Cosmochim. Acta* 15, 40 (1958); J. W. Morgan and J. F. Lovering, *Anal. Chim. Acta* 28, 405 (1963).

¹³The xenon calibration was improved by the use of xenon-gas standards.

¹⁴T. Kirsten, in *Potassium-Argon Dating*, edited by O. A. Schaeffer and J. Zähringer (Springer-Verlag, Berlin, Germany, 1966), p. 7.

$(\pi-\Sigma)_{I=0,1}$ AND $\bar{K}-N$ SCATTERING LENGTHS: EXPERIMENTAL AND THEORETICAL VALUES*

Frank von Hippel†

Institute of Theoretical Physics, Department of Physics, Stanford University, Stanford California

and

Jae Kwan Kim

Department of Physics, Harvard University, Cambridge, Massachusetts

(Received 4 April 1968)

We present experimental values of the $(\pi-\Sigma)_{I=0,1}$ and $\bar{K}-N$ scattering lengths derived from a coupled-channel effective-range analysis of the reactions $\bar{K}+N \rightarrow (\bar{K}+N, \pi+\Sigma, \pi+\Lambda)$, and theoretical values obtained by use of the soft-meson estimates of the scattering lengths in a role similar to that of known subtraction constants. Agreement of theory with experiment is within experimental error except for the $I=0$ $\bar{K}-N$ scattering length where the disagreement is by 40%.

A number of authors¹ have used partial conservation of axial-vector current (PCAC) and the charge commutation relations of $\text{SU}(3) \otimes \text{SU}(3)$ to derive, to first order in the meson four-momenta, the scattering amplitudes for pseudoscalar mesons on various targets. When these amplitudes are evaluated for elastic scattering at threshold, we obtain "soft-meson" estimates of s -wave scattering lengths. In the case of $\pi-N$ and $K-N$ elastic scattering, these estimates have been found to be in rough agreement with experiment (typically 30%).

In this paper we give new experimental values for the $(\pi-\Sigma)_{I=0,1}$ and $\bar{K}-N$ scattering lengths (see Table I). These scattering lengths have been obtained from a coupled-channel effective-range analysis² of the reactions $\bar{K}+N \rightarrow (\bar{K}+N, \pi+\Sigma, \pi+\Lambda)$ from 0 to 550 MeV/ c . The "soft-meson" estimates for these scattering lengths are also shown in Table I.³ There is obviously very little resemblance between the two sets of numbers.

We show below that this disagreement is due to a failure in the soft-meson approximation. It is not adequate to keep only terms of zeroth and first order in the meson momenta because the s -wave $\pi-\Sigma$ and $\bar{K}-N$ scattering amplitudes are large near their thresholds (resonant in the $I=0$ channel). This contrasts with the situation in $\pi-N$ and $K-N$ scattering where the s -wave unitarity cut is weak.

The PCAC, $\text{SU}(3) \otimes \text{SU}(3)$ results are far from useless, however, and it is still possible to test them. We show that the soft-meson results can be made to play a role similar to that of known subtraction constants, and that consequently the real part of each scattering length can be written approximately as a sum of the soft-pion scattering length and an integral over the imaginary part of the corresponding s -wave scattering amplitudes. Because of the subtractions, this integral is highly convergent and is, therefore, sensitive only to the amplitude in the region where it is known from Kim's effective-range expansion.²