The Variation of the Adiabatic Elastic Constants of Polycrystalline Ammonium Chloride with Temperature between 200°K and 273°K*

The data which follow were obtained with the dynamical method described in this journal by Balamuth[1] and Rose.[2] The form of specimen required by this method is a right circular cylinder 4.6 mm in diameter and about 3 cm long. Such cylinders are cut from a block of material prepared by compressing dry powdered ammonium chloride to 50,000 lb./in.² in a steel cylinder at room temperature. The result is a coherent isotropic array of microcrystals whose average linear dimensions are 5×10^{-3} cm. The densities of different blocks at 24°C vary slightly about the value 1.520 g/cm³, which is slightly less than the density of crystalline ammonium chloride as measured by Wulf and Cameron,[3] namely 1.527 g/cm³. It is found, however, that small changes in density are very nearly without effect on the *temperature variation* of the elastic constants, expressed in percent change per degree.

The temperature of the specimen is kept constant to 0.01°C during a measurement by manually controlling the temperature of an enveloping well stirred methanol bath. The temperature of the bath is measured with a platinum resistance thermometer, certificated by the National Bureau of Standards, used in connection with a Mueller bridge.

The curves of Fig. 1 show the observed variation of the adiabatic Young's and rigidity modulus with temperature through the phase transition which occurs at 242.8°K. The corresponding variations of the adiabatic compressibility and Poisson's ratio, as calculated from these data, are given in Fig. 2. The absolute values of the Young's modulus, rigidity modulus, compressibility and Poisson's ratio at 273.2°K are, respectively, 2.733×10^{11} dynes/cm², 1.096×10^{11} dynes/cm², 5.54×10^{-12} cm²/dynes, and 0.246.

The temperature of the transition point has been deduced from observations on the temperature variation of the specific heat at constant pressure by Simon, v. Simson and Ruheman,[4] and of the coefficient of thermal expansion by Smits and Gillavry[5] and Adenstadt.[6] Both quantities show a pronounced maximum at 242.8°K, which is exactly the temperature here found for the occurrence of the minimum value of Young's modulus and the maximum

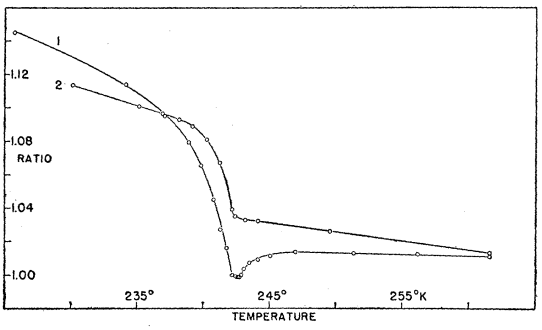

FIG. 1. The variation of the Young's modulus (curve 1) and the rigidity modulus (curve 2) with temperature near the transition point. The ordinates are the ratios of the values at the given temperature to the values at 273.2°K.

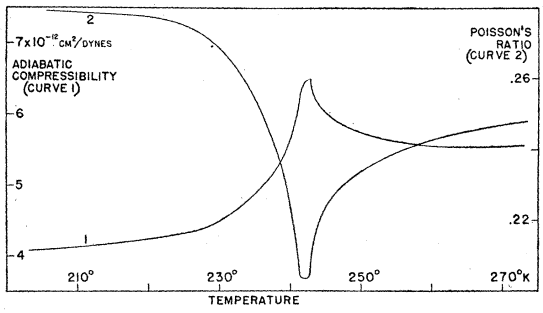

FIG. 2. The variation of the adiabatic compressibility (curve 1) and Poisson's ratio (curve 2) with temperature.

value of the compressibility. May[7] has computed the isothermal compressibility of ammonium chloride at 273°K from supplementary unpublished data supplied him by Bridgman.[8] The value given is 5.60×10^{-12} cm²/dynes.

The present measurements, together with those on the temperature variation of the coefficient of thermal expansion[5, 6] and on the heat of transition[9], indicate that the values of both c_p and the ratio c_p/c_v at the transition point may be much larger than has hitherto been believed. Measurements similar to the above on the variation of the isothermal moduli with temperature will settle the matter, and apparatus designed for this purpose is now under construction in this laboratory. Data will then be available for comparison with Fowler's theoretical calculation of the variation of c_v with temperature.[10]

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Long-Lived Radioactive Fe^{55}

In a previous paper[1] we have discussed the production of radioactive iron isotopes Fe^{53} and Fe^{59}, with half-lives 9 minutes and 47 days, respectively. Failure to observe any activity attributable to Fe^{55} led to the conclusion that its lifetime was either very short or extremely long. (The mass numbers of the stable iron isotopes[2] are 54, 56, 57 and 58.)

Continued observations over a period of 22 months on the chemically separated iron fractions from deuteron-bombarded iron samples show that the 47-day period is accompanied by a much weaker and very much longer-lived activity. That this cannot be ascribed to incompletely removed long-lived isotopes of cobalt[3] (~250 days) or manganese[4] (310 days), which are also produced by deuteron-bombardment of iron, is shown by the absence

of the very strong short-lived cobalt and manganese activities in these same iron samples. A particularly rigorous chemical separation, designed to remove all elements other than iron, was made on a more recently activated specimen; this precipitate, now 10 months old, has already departed from the 47-day period and is now going into one much longer.

These facts assure us that Fe^{55} has been formed through $Fe^{54}(d, p)Fe^{55}$ with the activity probably leading to stable Mn^{55} either by positron emission or by K-electron capture. An absorption curve on one of the old samples (unfortunately very weak) shows that the radiation has a soft component with half-thickness 3 to 5 mg/cm^2 of aluminum. This suggests the existence of an internally converted gamma-ray.

It is too early to give a reliable value for the half-life of this activity. The observed decay curves, which are still going through the transition interval from the 47-day period to the long period, set a lower limit of 1 year for the half-life. A comparison of the intensities of the 47 day and the longer activity (assuming equal cross sections for their production and equal ionizing powers for their radiations, and allowing for the abundances of the primary isotopes involved) predicts that the half-life will exceed 10 years.

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Stability of Uranium and Thorium for Natural Fission

The question of the occurrence of possible natural fission processes among the heavy nuclei is one of considerable importance to both nuclear physics and geophysics. The characteristics of the neutron-induced splitting of the uranium and thorium nuclei which have recently been studied so intensively by many workers seem to indicate that a naturally occurring explosion process probably would form radioactive isotopes of elements of medium atomic number with convenient half-lives. Further, the emission of neutrons by uranium in the artificial process suggests that neutron emission might accompany the natural process. Finally, natural fission might be detected by disparities between the results of the various calculations of the age of the oldest rocks based on the observed abundances of the elements and the intensities of the ordinary radioactive emissions from uranium and thorium.

It is the purpose of this letter to point out how well evidence on these three points excludes natural fission. In all of the numerous experiments which have been done on the neutron-produced activities of uranium and thorium control separations must have been made without neutron irradiation to exclude the possibility of contamination by natural activities. In order to have definite data for a typical calculation for this type of evidence the experiments[1, 2] on extraction of radioiodine from irradiated uranium were repeated with 0.90 mole of uranyl nitrate which had not been disturbed chemically for at least five years. The iodine, extracted by adding a little solid iodine to a slightly acidified aqueous solution of the uranyl nitrate, shaking with CCl_4, and precipitating AgI in a thin layer was counted in a screen wall counter so no beta-radiation harder than 20,000 electron volts would have been missed. The result was zero within 20 counts per minute. If it is assumed that each ten explosions produced at least one radioactive iodine on the average this result requires that the half-life for natural fission be at least as long as 10^{14} years. Similar results must apply for the other elements which have been separated, so we can conclude that natural fission to produce any of the radioactive elements found in the artificial process must correspond to a half-life of the order of or greater than 10^{14} years. A similar test was performed on thorium with the same results.

In order to investigate possible natural neutron emission from uranium and thorium a BF_3-filled counter was surrounded with paraffin and standardized with 200 mg of $RaBr_2$ mixed with Be powder. Seven and one-half moles of uranium salts were found to give less than two counts per minute whereas the Ra-Be source in the same position gave 4600 counts per minute. If it is assumed that the Ra-Be source gives 17,000 neutrons per second per millicurie,[3] the half-life for fission must be at least as large as 10^{14} years if at least one neutron were emitted per explosion. Similar measurements on a smaller amount of thorium gave a lower limit of 5×10^{13} years.

The agreement between the uranium and thorium age determinations on old rocks and the agreement of these with other estimates of the age of the oldest strata, e.g., the Sr^{87} : Rb^{87} ratio found by Hahn, Strassmann and Walling[4] in lepidolite from the Canadian shield indicate that the age of the earth is at least 2×10^9 years and that no fission process with a half-life less than 5×10^9 years occurs, unless uranium and thorium have been or are now being generated by some terrestrial process. In the absence of any evidence for such a generation we must conclude that the half-life for any possible fission exceeds 5×10^9 years.

Consideration of all these points leads to the conclusion that no fission similar to those induced by neutron absorption can occur naturally for either uranium or thorium at a rate greater than that corresponding to a half-life of 10^{14} years and that no natural fission of any sort has a life less than 5×10^9 years.

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