OKLAHOMA GEOLOGICAL SURVEY

CHARLES J. MANKIN, Director



URANIUM AND NATURAL RADIOACTIVITY IN OKLAHOMA

Ву

MATTHEW W. TOTTEN AND ROBERT O. FAY

(Text to accompany map GM-25)

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MAP SHEET

(Separate, in envelope)

Map of Oklahoma showing localities of reported uranium and radioactivity values

URANIUM AND NATURAL RADIOACTIVITY IN OKLAHOMA

MATTHEW W. TOTTEN¹ AND ROBERT O. FAY²

Abstract—Uranium (U) is the heaviest natural element known. It is radioactive, giving off alpha, beta, and gamma radiation, breaking down into 35 lighter elements. Helium, radium, and radon gas are some of the common daughter products of uranium. Each element can be identified by the type and amount of radiation emitted, using 16 different methods of analysis.

Uranium occurs in nature in combination with oxygen and other elements; it does not occur in nature as a metallic element. Uranium occurs in all major types of rocks and in oil and water.

Carbon, hydrogen sulfide, and phosphate commonly attract uranium for precipitation.

Commercial uranium ore is generally minable if the $\rm U_3O_8$ content is greater than a cutoff grade of 0.1 percent or 1,000 parts per million or 2 pounds per ton, valued at \$20.00 to \$46.00 a pound. Some cutoff grades in the western United States are as low as 0.03 to 0.06 percent, with the average being less than 0.1 percent. In Oklahoma, only one place had minor commercial ore, at Cement, Caddo County, where 13 tons of carnotite ore averaging 2.2 percent $\rm U_3O_8$ was mined from the Rush Springs Sandstone in 1956.

Almost all uranium occurrences in Oklahoma are noncommercial, most being low-grade or small ore bodies or simply being reports of radioactivity in igneous and sedimentary rocks, oil, and water.

Approximately 400 radioactive localities were noted in Oklahoma prior to 1980.

Introduction

The purpose of this report is to summarize previous knowledge of uranium and radioactivity in Oklaho-

ma with respect to geology.

Uranium (U) is a radioactive, unstable white metal with an atomic number of 92 (92 protons and 92 electrons) and an atomic weight of 238 (92 protons and 146 neutrons). It is the heaviest naturally occurring element and occurs in nature in combination with other elements. Two other uranium isotopes occur in nature with U^{238} , in approximate proportions of U^{238} (99.28 percent), U^{235} (0.71 percent), and U^{234} (0.0054 percent). Uranium has a density of 18.97 to 19.04 grams per cubic centimeter.

Uranium was discovered in 1789 by a German chemist, M. S. Klaproth, who named it in honor of the planet Uranus, which was discovered 8 years previously. In 1841, E. M. Peligot first chemically isolated uranium. In 1896, the French scientist Henri Becquerel discovered the radioactive property of uranium. In 1898, Marie and Pierre Curie and G. Bémont discovered radium while working with pitchblende (UO2 variety). In 1939, Enrico Fermi suggested that when uranium nuclei are split under neutron bombardment, other neutrons might be released in a fission reaction. A little later it was proved that the U^{235} isotope is the one that is split in the fission process. On December 2, 1942, Fermi and others produced the first self-sustaining nuclear chain reaction at the University of Chicago. On July 16, 1945, the first atomic bomb was exploded. In 1955, nuclear power was first used for propulsion, in the submarine U.S.S. Nautilus. On December 2, 1957, the first nuclear-powered electrical-generating station began operation at Shippingport, Pennsyl-

The nuclear-decay scheme of U²³⁸ and daughter products is shown in table 1, and that of U²³⁵ is shown in table 2. Rutherford and Royds demonstrated in 1907 that alpha particles are helium (₂He⁴) nuclei,

with 2 protons and 2 neutrons, and thus all nuclear reactions involving the loss of alpha particles are responsible for the origin of helium. The alpha particles gain two electrons each to become a neutral helium atom. From 1899 to 1919, Rutherford and his associates discovered that three types of radiation were associated with radioactive decay, which they termed alpha, beta, and gamma, resembling anode, cathode, and x-rays respectively.

Alpha rays are positively charged particles of mass number 4 and atomic number 2, slightly deflected by electrical and magnetic fields, with slight penetrating power. Beta rays are electrons, negatively charged, derived when an electron is lost and a neutron becomes a proton strongly deflected by electrical and magnetic fields, and 100 times more penetrating than alpha rays. Beta decay causes the atomic number to increase one unit. Gamma rays are photons of electromagnetic radiation, undeflected by electrical and magnetic fields, with wavelengths of 10⁻⁸ to 10⁻⁹ cm, and 10,000 times more penetrating than alpha rays. Half-life is defined as the time required for the disintegration of half of the atoms in a sample of a specific radioactive element.

In 1974, the U.S. Nuclear Regulatory Commission estimated that the reasonably assured uranium reserves in the United States were 277,000 short tons of $\rm U_3O_8$ contained in 129×10^6 (1.29 billion) tons of ore with an average grade of 0.21 percent $\rm U_3O_8$ and recoverable at a cost of \$8.00 per pound or less. The potential reserves at \$8.00 per pound $\rm U_3O_8$ were estimated at 450,000 tons. In 1980, the U.S. Department of Energy estimated the reserves of the United States to be 645,000 tons of $\rm U_3O_8$ at a forward-cost of \$30.00 per pound, or 936,000 tons at \$50.00 a pound, or

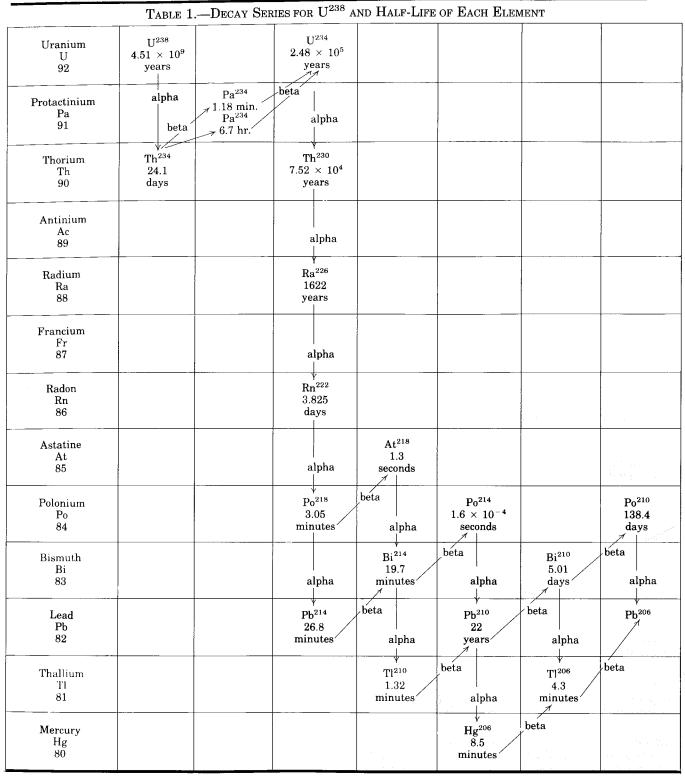
1,122,000 tons at \$100.00 a pound.

Uranium and other radioactive minerals occur in many places in Oklahoma. The reported occurrences are generally noncommercial except for a few deposits. The only reported minor commercial uranium in Oklahoma is that at Cement, Caddo County, where about 13 tons of carnotite ore, averaging 2.2 percent U₃O₈, was mined from a vein in the Rush Springs Sandstone (Permian) in the summer of 1956 (map no. 46). This paid off at \$6.00/pound only be-

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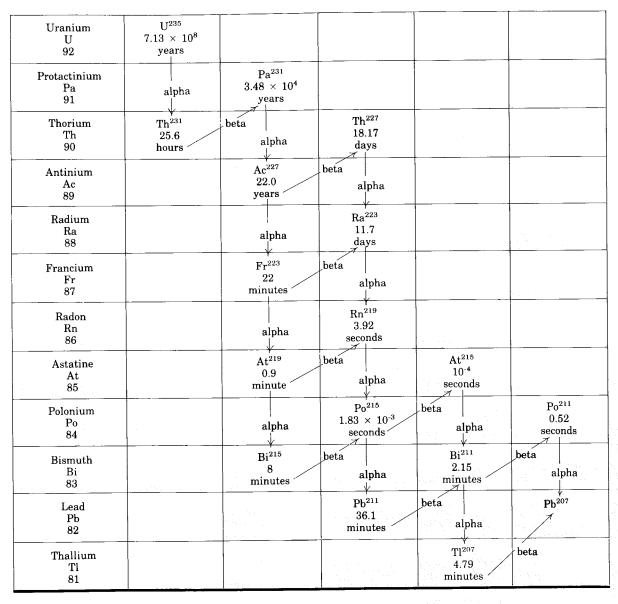
cause the U.S. Atomic Energy Commission paid the shipping charges. All other reported occurrences are simply reported values of uranium analyses of oils, asphaltites, asphaltic sandstones, asphaltic pellets, brines, sump-pit material, black shales, phosphatic nodules, igneous rocks, sandstones, red beds, fossil wood, copper deposits, radium-spring water, saltspring water, fresh water, or well water. Helium is reported in natural gases. Much of the radioactivity may be due to radioactive potassium in shales or to daughter products from uranium, such as radon or radium. The various analyses and sources of data are

listed in the tabulation on the accompanying map. The tabulation lacks the recent NURE (U.S. Department of Energy's National Uranium Resource Evaluation) data for the Tulsa, Enid, Clinton, and Lawton $1^{\circ} \times 2^{\circ}$ sheets and lacks water and gamma-ray analyses by the Oklahoma State Department of Health as well as airborne gamma-ray-spectrometer values.

Methods of Analyses

About 16 different analytical techniques are used to detect and measure uranium (table 3). The com-

Table 2.—Decay Series for U^{235} and Half-Life of Each Element



mon methods utilize a geiger counter, nucleometer, scintillometer, and gamma-ray spectrometer. Autoradiography is also used. A good treatment of these subjects is given by Harold Wollenberg (1977, p. 5–35), Chapter 2, Radiometric Methods, in *Nuclear Methods in Mineral Exploration and Production*, edited by Jerome Morse (Elsevier Scientific Publishing Co., New York, Developments in Economic Geology, 7).

The geiger counter and nucleometer have a gasionization chamber that detects alpha and beta particles by their conductivity, which is recorded generally in counts per second (cps). A scintillometer has a fluorescent screen such as zinc sulfide or sodium iodide that flashes light when struck by gamma rays, and these flashes are recorded in milliroentgens per hour (mr/hr), or counts per minute or second (cpm)(cps), or million electron volts (MeV). A gamma-ray spectrometer has a defective crystal such as thallium-doped sodium iodide or lithium-drifted germanium that can detect different wavelengths or energy levels of gamma rays, and these are recorded

accordingly for each source, such as uranium, thorium, and potassium.

Autoradiography is the study of decay particle tracks directly from a radiometric source. (1) Alpha autoradiography is the study of alpha tracks on a photographic emulsion, after placing a thin section of a uranium-bearing rock against the emulsion for 10 days or more. (2) Fission-track autoradiography is the study of etched tracks in muscovite mica or polycarbonate plastic Lexan or Makrofol, placed against the thin section and against standards, and bombarded by neutrons in a reactor for about 3 minutes. (3) Radon autoradiography is the study of alpha tracks from radon. Ra²²² is the gaseous decay product of U²³⁸, with a half-life of 3.8 days. Ra²²² decays to Pb²¹⁰ with the emission of 3 alpha particles of 5.48, 6.0, and 7.69 MeV energy levels. Plastic cups with alpha-track detectors, such as a silicone conductor, are inverted several feet down in the soil and left for several weeks. The alpha particles from Ra²²² are recorded on the detector, which is etched, and the number of tracks is noted. One variation is the place-

TABLE 3.—ANALYTICAL TECHNIQUES FOR URANIUM

Name	Principle ^a	Approximate Detection Limit, (µg/g)	Probable Error
Fission track	NA, ND	10-4	5.–10.
Isotope dilution	MS	10-4	0.5-5.
Neutron activation	NA, ND	10-4	25.
Delayed neutron counting	NA, ND	10^{-3}	3.
Emission spectroscopy	ET	5×10^{-2}	110.
Fluorescence (ultraviolet			
and x-ray)	ET	10-1	550.
Chromatography	WC	10-1	10.
Gamma-ray spectrometry	ND	10-1	10.
Autoradiography	ND	1.	110.
Titrimetry	WC	1.	0.5-5.
Colorimetry	WC	10.	13.
Atomic absorption	ET	5×10^{1}	2.–5.
Alpha counting			
(electronic)	ND	5×10^{1}	110.
Polarography	EC	10^{2}	25.
Potentiometry	EC	2×10^2	1.–5.
Gravimetry	WC	5×10^4	0.1–2.

^a ND, nuclear decay; ET, electron transitions; WC, wet chemical; EC, electrochemical; NA, neutron activation; MS, mass spectrometry.

From Haglund, D. S., Uranium: Element and Geochemistry, in *The Encyclopedia of Geochemistry and Environmental Sciences* by R. W. Fairbridge (editor). Copyright © 1972 by Dowden, Hutchinson & Ross, Inc., Stroudsburg, Pennsylvania. Reprinted by permission of the publisher.

ment of an electronic recorder in the hole to register integrated counts instead of etching the surface.

Definitions

Some common terms used in analyses and measurements are listed in alphabetical order.

API units-100 API units = 17 ppm eU.

c—means chemically determined. Chemical analyses are expressed as %cU or in parts per million (ppm) or parts per billion (ppb). $1\times(\%$ or ppm)cU = $0.85\times(\%$ or ppm)cU₃O₈. Also, 1% U₃O₈ = 20 lb U₃O₈ per ton of ore, or 0.1% U₃O₈ = 2 lb U₃O₈/ton of ore. Thus 85 lb uranium metal = 100 lb U₃O₈.

Curie (Ci)—A curie is the official unit of radioactivity in the field of radiation dosimetry, defined as 3.70×10^{10} disintegrations per second. One gram of radium²²⁶ has slightly less than 1 curie of radioactivity. 1 microcurie (μ Ci), or one millionth of a curie, = 1×10^{-6} curie. 1 nanocurie (nCi), or one billionth of a curie, = 1×10^{-9} curie. 1 picocurie (pCi), or one trillionth of a curie, = 1×10^{-12} curie. The maximum safety standard for radioactivity from Ra²²⁶ in drinking water has recently been determined to be 5 picocuries per liter (pCi/L) by the U.S. Environmental Protection Agency (EPA). The standard for gross alpha is 15 pCi/L. One municipal water supply in the State exceeds the permissive level of Ra²²⁶, but that city is taking action to eliminate the excessive level.

Erg—A unit of energy. 1 erg = 10^{-7} joule. 1 foot-pound = 13,560,000 ergs. An erg is the work done when a force of 1 dyne is applied through a distance of 1 cm.

eU—means possible equivalent uranium. Percent eU is a measure of radioactivity reported as the concentration of uranium in equilibrium that would produce the measured radioactivity. Thus ${\rm Ra}^{226}=0.1\%$ eU indicates that the sample contains the amount of ${\rm Ra}^{226}$ that would be in equilibrium with 0.1 percent uranium

Electron volt (eV)—An electron volt is a unit of energy and equals 1.602×10^{-12} erg. A million electron volts is MeV. The unit is used in connection with ionization or excitation of atoms or molecules by gamma rays or other types of radiation. Each radioactive element in a decay series has a particular gammaray-emission wavelength or energy level. The common ones that are recorded are 1.46 MeV for potassium (K⁴⁰), 1.76 MeV for bismuth (Bi²¹⁴), and 2.62 MeV for thallium (Tl²⁰⁸). Gamma-ray spectrometers are electronically adjusted to record gamma-ray energies from 0.2 to 4 MeV, which includes most of the radioactive-decay series for uranium and thorium.

Gamma-ray log—A down-hole log of total gamma radiation from rock. Hyden and Danilchik (1962), using gamma-ray logs, used a conversion factor of 0.0007 percent eU per 1 inch of deflection on a 10 inch sensitivity scale, with an error of \pm 0.001 percent eU, to relate gamma rays to percent eU. If the gamma rays are not related to uranium, then percent eU may not be related to uranium.

mer—A mer is that amount of benefit required to justify an exposure to 1 rem.

microgram (μ g)—one millionth of a gram, equal to 1×10^{-6} grams. 1 microgram per liter (μ g/L) = 1 part per billion (ppb) = 0.0000001 percent cU.

milligram (mg)—one-thousandth of a gram, equal to 1×10^{-3} grams. 1 milligram per liter (mg/L) = 1 part per million (ppm) = 0.0001 percent cU.

rad—A rad is 100 ergs of energy per gram of absorbed ionized radiation. The EPA safety standard is 0.17 rad per person per year.

rem—A rem is the radiation damage to a human equal to 1 roentgen of 200 kilovolts x-radiation.

roentgen—A roentgen is the international unit of exposure or quantity or dose for x-rays, gamma rays, alpha particles, beta particles, protons, and neutrons. It is defined as the quantity of gamma or x-rays that will produce one electrostatic unit of electricity of either sign in 1 cc of dry air at 0°C and standard atmospheric pressure, as a result of ionization. On any scalar reading, 1 milliroentgen per hour (mr/hr) = 500 counts per minute (cpm), or 1 count per minute = 0.002 mr/hr.

Geochemistry

The uranyl ion $(\mathrm{UO}_2)^{+2}$ is mobile under oxidizing conditions and is carried in solution as a uranyl dicarbonate complex or as a uranyl tricarbonate complex, according to Garrels, Hostetler, and Christ (1957, p. 551). The uranium is precipitated under reducing conditions, forming uraninite and other minerals (fig. 1).

Breger and Deul (1956, p. 505–509) suggest three ways in which uranium may be deposited: (1) organic material reduces the soluble uranyl ion to an insoluble uranous ion; (2) the sulfide ion is a reducing agent; and (3) carbonaceous material, like coal, may adsorb uranium from ground water, forming insolu-

ble uranyl humates. Uranium may be carried as a sulfate complex, $\rm UO_2SO_4$, in solutions with pH equal to 6.5 or less.

McKelvey, Everhart, and Garrels (1956, p. 41–53) stated that the ultimate source of uranium may have been devitrified volcanic ash, weathered igneous rocks, and hydrothermal solutions. The uranium was probably adsorbed by humates and other organic compounds that came in contact with uranium in solutions migrating through rocks. Apatite in phosphorites probably chemically adsorbs uranium. Phosphate complexes strongly with uranium and is important in keeping U⁺⁶ in solution. Sedimentary rocks contain more vanadium than igneous rocks. Oil is not a primary source of uranium.

In Oklahoma, the Precambrian Spavinaw Granite contains 5 ppm uranium, and the Cambrian Wichita Granite Group and Carlton Rhyolite contain an average of 3 ppm uranium. The range of uranium in the Carlton Rhyolite is 2 to 16 ppm. Pegmatites in the Quanah Granite range from 40 to 100 ppm in uranium content. About one-half of the uranium is tightly locked in zircons and is unavailable to solutions.

Helium is a byproduct from the radioactive breakdown of uranium in arkosic sediments, reaching natural gas by bubble diffusion. The Morrowan Keyes sandstone in Cimarron County contains more than 2 percent helium in the natural gas.

High uranium concentrations (up to 20,460 ppm) have been reported from Pennsylvanian, Permian, and Triassic red beds. These occurrences are associated with hydrocarbon halos over known oil fields, with fossil wood, or with a roll front (an area of reduction and concentration of uranium below the ground-water table).

For further details, see other chapters on uranium analyses from water, oil, black shales, coal, and red beds.

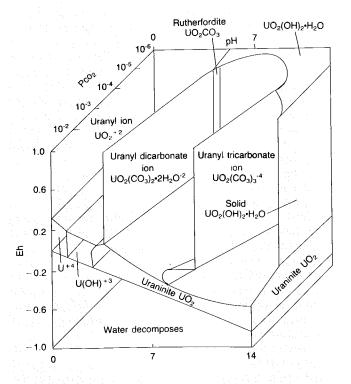


Figure 1. A portion of the system U-H₂O-CO₂ at 25°C.

Mineralogy

The common uranium minerals reported from Oklahoma, mostly from red beds, are carnotite $K(UO_2)(VO_4)_2 \cdot 1-3H_2O$ and tyuyamunite $Ca(UO_2)_2$ $(VO_4)_2 \cdot 7-10H_2O$. Other minerals, mostly found in Permian red beds, are autunite $Ca(UO_2)(PO_4)_2 \cdot 8-12H_2O$, bayleyite $Mg_2(UO_2)(CO_3)_3 \cdot 18H_2O$, coffinite $UO_2 \cdot SiO_2$, novacekite $Mg(UO_2)_2(AsO_4)_2 \cdot 10H_2O$, torbernite $Cu(UO_2)(PO_4)_2 \cdot 8H_2O$, uraninite $UO_2 \cdot 2UO_3$, and uranophane $Ca(UO_2)_2Si_2O_7 \cdot 6H_2O$.

The uranium in most of the black shales, brines, water, oil, asphaltites, coal, and phosphatic nodules may be attached to other organic or mineral matter and may not be in a distinctive uranium mineral.

Coffinite and uraninite have been reported to occur in asphaltic pellets. Novacekite occurs in the Post Oak Conglomerate (Permian) of the Wichita Mountains, and it could have been recently formed. Carnotite was the common ore at Cement, Caddo County. Torbernite is associated with copper deposits. Tyuyamunite and carnotite are the common minerals in the basal Doxey Shale (Permian) of western Oklahoma. Autunite and bayleyite occur with fossil plant remains in red-bed sandstone channels.

Water Analyses for Uranium

Fresh water.—In the Oklahoma Panhandle, fresh water generally contained less than 30 ppb uranium, and municipal water contained less than 14 ppb uranium. Landis (1957, table 37; 1961, p. 223-258) summarized knowledge of uranium in fresh water in the Panhandle. Waters in the Ogallala tuffaceous fluviatile sandstones (Tertiary) average 6.7 ppb uranium, in Cretaceous shales 20.4 ppb, and Cretaceous terrestrial and near-shore sandstones, such as the Dakota, 10.2 ppb uranium. One water sample from the Jurassic Exeter Sandstone contained 4 ppb uranium. Two water samples from the Triassic Dockum Group averaged 38 ppb uranium. Six water samples from the Permian Doxey and Cloud Chief Formations contained 80 to 160 ppb uranium. The Permian Rush Springs Sandstone contained as much as 800 ppb uranium at Cement, according to Olmsted (1975).

In northeastern Oklahoma, many fresh-water and brine springs flow from the Mississippian Boone Group, or from beds above and below, such as the salt springs at Nicut, Sequoyah County, which flow from the base of the Pennsylvanian Hale Sandstone. Some of these springs are radioactive. Siebenthal (1908, p. 226-227) mentioned that the radium springs at Claremore are from well water more than 1,100 feet deep to the lower Chattanooga black shale (Devonian) and possibly the Roubidoux Sandstone (Ordovician). The water wells at Kansas, Delaware County, are drilled to the Roubidoux Sandstone, and the cuttings have sand grains coated with a uranium mineral resembling carnotite. It is possible that the radioactivity in the water at Welch, Craig County, and Afton, Ottawa County, is coming from uranium minerals in the Roubidoux Sandstone. The Roubidoux in this area needs to be studied with respect to radioactivity.

Brine water.—Hyden and Woods (1955, p. 179–183) stated that oil-field brines from Nowata, Nowata County, range from 6 ppb to less than 1 ppb uranium. Branson, Burwell, and Chase (1955, p. 19) stated that these brines are from the Bartlesville and Burgess sandstones (Desmoinesian). Pierce, Mytton, and Gott (1956, p. 527–532) stated that uranium as high as 0.2 ppm is present in the brines of the Amarilo—Wichita Uplift.

Bloch (1979a, p. 177–182) mentioned that many oil-field brines may be high in radium-226 and low in uranium-238, owing to the geochemical environment of petroleum reservoirs and to the chemical properties of the radium. Many of the rocks containing the brines may be low in uranium-238.

Bell (1960, p. 56) mentioned that sea water contains 0.37 to 3.5 ppb uranium, varying with salinity, depth, and position with respect to shoreline. The average concentration of uranium in sea water is 3.3 ppb, according to Rodman and others (1979).

Oil and Asphaltic-Rock Analyses for Uranium

Oil.—Hyden (1956, p. 511-519) stated that uranium in oils in general is not preferentially distributed with respect to age, lithology, rock type, or type of oil. Vanadium and nickel are present in oils as porphyrin complexes, but uranium is not, according to Hyden. (A porphyrin is an organic complex derived from hemoglobin and chlorophyll, according to Hyden.) Bell (1960, p. 45–65) mentioned that sea water contains three times more uranium than paraffin-based oils and that petroleum is a poor source for uranium but may help to concentrate uranium. The uranium may be attached to the asphaltene portion or to colloidal organic material in the oils. Natural paraffin oils contain 1 ppb or less of uranium. Hydrogen sulfide is abundant in crude oils and is an excellent reducing agent. Bell reported that 37 crude-oil samples from Ordovician through Permian rocks contained 0.1 to 248.4 ppb uranium. Hyden and Woods (1955, p. 183) mentioned that uranium in oils is probably not a uranium-porphyrin complex because of the low content of nitrogen in the oil (0.018 to 0.48 percent) compared to the uranium content of the ash in the oil (1 to 75 ppm), with each sample showing a negative correlation coefficient.

Asphaltic rocks.—Bell (1960, p. 45–65) analyzed 45 asphaltic rocks from Ordovician through Permian ages in Oklahoma . The oil portions ranged from 0.58 to 12.28 percent, and the uranium in the oil portions ranged from 0.004 to 17.1 ppm uranium, with a few ranging up to 140 ppm. The bitumens in the asphaltic rocks averaged 1,000 ppb uranium. The heavier the gravity of the extracted oil, the greater the uranium content. Hail and others (1956, p. 521–525) mentioned that the uranium is concentrated as organouranium complexes in the asphalt portion of the host rock. The amount of uranium is probably related to the source sediments of the asphalts or to the rocks through which the oil migrated.

Asphaltites and asphaltite pellets.—Beroni (1956) mentioned that asphaltite pellets contain smaltite, uraninite, coffinite, and trace elements similar to those of crude oils. Hill (1954, p. 1377) suggested that asphaltite pellets with smaltite and uraninite may

be derived from asphalt seeps. Hill (1953, p. 200–204) suggested that the original source for the uranium in asphaltite pellets in Permian red beds around the Wichita Mountains may have been from the Wichita igneous rocks. Pierce (1954, p. 274–276) studied trace metals in asphaltites in Permian red beds around the Wichita–Amarillo Uplift. He found that the trace metals occur in inclusions in arsenide and sulfide minerals in the asphaltite. The inclusions were apparently formed from solidification of the asphaltite, which is evidence that the asphalt passed through a liquid phase.

Hill (1957, p. 1-7) stated that carbonaceous nodules in Permian red beds around the Wichita Mountains contain 3,700 to 8,600 ppm uranium and have trace metals similar to those of crude oil in the area. These nodules may be the product of microbiological oxidation of petroleum.

Red-Bed Analyses for Uranium

Fisher and Stewart (1960, p. 42–44) stated that uranium in Permian red beds was probably precipitated from circulating ground water under reducing conditions. Three main types of deposits are (1) those formed beneath the ground water table, or roll-front deposits; (2) those formed around oil fields where H₂S is prevalent; and (3) those formed around organic material such as fossil wood. The second type, or the hydrocarbon-halo type, has received much attention, especially the Cement deposit in Caddo County.

Ferguson (1979, p. 206) suggested that connate brines moving from the basin toward the margins carry oil droplets with the water. The connate water mixes with meteoric water moving down the margins of the basin. The oil accumulates in favorable reservoirs, and the uranium is carried in low concentrations in the water. Later movements along faults facilitate the release of hydrocarbons and H₂S, which may react with certain cements and mineral-rich waters to produce pyrite and new calcite cement. Some plausible reactions are $C_nH_m + 3CaSO_4 \rightarrow$ $3\text{CaCO}_3 + 3\text{H}_2\text{S} + \text{CO}_2 + \text{H}_2\text{O}$, and $2\text{H}_2\text{S} + \text{Fe}_2\text{O}_3 \rightarrow$ FeS₂ + FeO + 2H₂O. Any uranium in the waters would be deposited. Bell (1960, p. 59) stated that no relationship exists between oil and uranium in the Cement Field and that the oil was not the source for the uranium. Olmsted (1975, p. 1–116) suggested that the uranium at Cement, in the Permian Rush Springs Sandstone, was probably derived from ground water moving through the sediments. Many of the sediments were probably derived from erosion of igneous rocks in the Wichita Mountains. Olmsted follows Garrels and others (1957, p. 550) in suggesting that the uranyl ion is probably carried as a uranyl dicarbonate complex in ground water under oxidizing conditions and deposited under reducing conditions around the halo of the Cement Field. The main reducing agent was probably H₂S. Some groundwater samples at Cement contained 800 ppb uranium, but these may be related to a high content of total dissolved solids.

Donovan (1974, p. 429–440) and Donovan, Friedman, and Gleason (1974, p. 351–354) showed that the hydrocarbon alteration at Cement extends to a depth of 2,500 feet. The carbonate cement has a carbon

isotopic composition ranging from -5.47 to -39.22 per mil, using a PDB-1 standard (Peedee belemnite from the East Coast Cretaceous). The C^{13} increases systematically relative to the C^{12} away from the crest of the Cement Anticline. Thus, the flanks of the anticline are isotopically heavier in carbon. The oxygen isotope ratios in the carbonate cement range from $\delta O^{18} = 22.5$ to 36.5 per mil, showing an increase of the heavy oxygen isotope toward the anticlinal crest, using Standard Mean Ocean Water (SMOW) as the oxygen standard. Thus Donovan and his associates concluded that the alteration at Cement was due to microseepage of hydrocarbons and H_2S .

The red-bed sandstones and shales in selected beds contained 30 to 16,100 ppm uranium. The vein at Cement contained 20,460 ppm uranium. Carbonaceous nodules contained 3,700 to 8,600 ppm uranium. Well water contained 7.4 to 160 ppb uranium

in Permian red beds.

Black-Shale and Phosphatic-Nodule Analyses for Uranium

The black shales in Oklahoma contain 6 to 900 ppm uranium but generally range below 200 ppm uranium. The higher values are associated with

phosphate nodules.

McKelvey and Nelson (1950, p. 35–53) stated that many marine black shales contain 100 to 200 ppm uranium. The nature of the uranium mineral in phosphatic nodules is unknown. McKelvey, Everhart, and Garrels (1956, p. 41–53) mentioned that uranium is precipitated in phosphorites by chemical adsorption of apatite. Russell (1945, p. 1470–1493) stated that marine oil shales have high radioactivity and that other types of organic sediments have intermediate radioactivity.

Devonian shales.—Landis (1955, p. 249–252) stated that the lowermost part of the Chattanooga Shale of Kansas has the greatest radioactivity and that the radioactivity increases southward toward Oklahoma. The radium springs at Claremore, Rogers County, Oklahoma, may come from well water from the Chattanooga (these springs are not natural). The Chattanooga of Oklahoma contains 10 to 130 ppm uranium, with higher values being associated with phosphate nodules.

Swanson (1954, p. 160–164) collected 75 samples of Woodford Shale, with the average uranium content being 30 ppm. Later, Swanson (1956, p. 451–456) stated that the Woodford Shale averaged 50 ppm in

uranium content.

Pennsylvanian shales.—Danilchik and Hyden (1957, p. 427–428) stated that Pennsylvanian black shales average 20 to 90 ppm uranium, and carbonaceous black shales average 10 to 20 ppm uranium. The phosphatic nodules in the shales range in content from 90 to 230 ppm uranium and seem to have concentrated the uranium.

Hyden and Danilchik (1955, p. 253–257) stated that the Excello Shale of northeastern Oklahoma ranges from 5 to 100 ppm in uranium content. Phosphatic nodules from the shale contained as much as 250 ppm uranium.

Coal and Coaly-Rock Analyses for Uranium

Breger and Deul (1956, p. 505–509) suggested that carbonaceous material, like coal, may absorb uranium from ground water, forming insoluble uranyl humates. Russell (1945, p. 1470–1493) stated that coals have abnormally low radioactivity and that other types of organic sediments have intermediate radioactivity. Vine (1962, p. 113–170) mentioned that the average uranium content of coaly carbonaceous rocks is less than 1 ppm but that some coaly rocks contain 50 ppm uranium.

Danilchik and Hyden (1957, p. 427–428) showed that Pennsylvanian carbonaceous black shales in Oklahoma average 10 to 20 ppm in uranium content.

Three Desmoinesian coal beds in northeastern Oklahoma were found to contain 10 to 40 ppm uranium.

Stratigraphic Uranium Analyses

Precambrian and Cambrian basement igneous rocks.—The Precambrian Spavinaw Granite contains about 5 ppm uranium. The Cambrian Carlton Rhyolite and Wichita Granite Group average 3 ppm uranium content, with the former ranging from 2 to 16 ppm uranium content. Pegmatite dikes of the Quanah Granite contain 40 to 100 ppm uranium.

Ordovician.—The Simpson and Viola oils contain 0.05 to 3 ppb uranium. The Roubidoux Sandstone and higher Arbuckle units may be a partial source for radioactive well water enriched with radium in

northeastern Oklahoma.

Silurian—Devonian.—The Arkansas Novaculite in the Ouachita Mountains contains 10 to 30 ppm uranium.

The Woodford Shale in the Arbuckle Mountains contains 6 to 127 ppm uranium, with the higher values being associated with phosphate nodules.

The Chattanooga Shale of northeastern Oklahoma contains 10 to 130 ppm uranium, with the higher values being associated with phosphate nodules. Radium springs and radioactive water may originate in the Chattanooga, especially the radium springs from well water at Claremore, Rogers County.

Mississippian.—The lower Stanley Shale near the Potato Hills in the Ouachita Mountains contains

about 10 ppm uranium.

The Delaware Creek Shale in the Arbuckle Mountains and outer belt of the Ouachita Mountains con-

tains 10 to 24 ppm uranium.

The Overbrook asphaltic sandstone of the Goddard Formation near Woodford, Carter County, contains 0.7 to 11 ppm uranium in the oil portion, with the higher amounts being in the heavier asphaltene portions.

Pennsylvanian.—Morrowan: The Jackfork asphaltic sandstone near Page, Le Flore County, contains 0.7 ppm uranium. The Keyes sandstone in the Keyes Field of Cimarron County contains 2.1 to 2.3 percent helium in natural gas from 4,630 to 4,873 feet deep.

Desmoinesian: The asphaltic sandstones contain 0.03 to 0.4 ppm uranium in the oil portion. The crude oils contain 0.1 to 28 ppb uranium. The black shales contain 1 to 600 ppm uranium. The Drywood, Tebo,

and Weir-Pittsburg coals contain 20, 40, and 10 ppm

uranium, respectively.

Missourian, Virgilian, and Gearyan: The asphaltic sandstones contain 0.2 to 140 ppm uranium in the oil portion. The crude oils contain 0.4 to 51 ppb uranium. The black shales contain 10 to 900 ppm uranium. Helium in the Ada group is 1.01 to 1.19 percent of the natural gas. Copper-bearing sandstones of the Wreford Formation in Pawnee County contained 800 to 10,700 ppm uranium (Lee Uto prospect).

Permian.—The asphaltic sandstones contain 0.6 to 100 ppm uranium in the oil portion. The crude oils contain 0.3 to 240 ppb uranium. The red-bed sandstones and shales, usually with copper mineralization, contain 30 to 16,100 ppm uranium, with the Cement deposit in Caddo County containing 20,460 ppm uranium. Carbonaceous nodules in red beds contain 3,700 to 8,600 ppm uranium. Well water in red beds contains 7.4 to 160 ppb uranium.

Triassic.—Well water in the Dockum Group of the Oklahoma Panhandle contains 38 ppb uranium. A lease play southwest of Boise City, Cimarron County, is supposed to be related to uranium in the Dockum

Group.

Jurassic.—Well water in the Exeter Sandstone of Cimarron County contains 4 ppb uranium. The Morrison Formation contains 700 to 2,600 ppm uranium in shales but contains 24 to 80 ppm in cherts and 8 to 14 ppm uranium in sandstones.

Cretaceous.—The Antlers Formation, south of the Arbuckle Mountains, contains 2.9 ppm uranium in asphaltic sandstone and 7,670 ppm uranium in

carbonized wood in sandstone.

The Purgatoire and Dakota Sandstones of the Oklahoma Panhandle contain 7 to 8.5 ppb uranium in well water and municipal water, and 25.5 ppb uranium in spring water.

The Upper Cretaceous shales in the Panhandle contain 2.3 ppb uranium in well water and 14.8 ppb

uranium in spring water.

Pliocene.—Fresh water in the Ogallala Formation contains less than 1 to 14.5 ppb uranium, with the higher readings from lake water. Municipal water from the Ogallala averages 6.5 ppb uranium.

Pleistocene and Holocene.—Fresh water from wells and springs in alluvium, terrace deposits, and dune sands and from streams, reservoirs, and lakes ranges from less than 1 ppb to 31.9 ppb in uranium content, with municipal water ranging from 4 to 14 ppb in uranium content.

Economic Geology

Uranium ore has commercial value when the ore can be extracted at a profit. At present, the uranium ore mined in the western United States is 0.03 to 0.1 percent, or 300 to 1,000 ppm, or more U_3O_8 , valued at \$20.00 to \$46.00 a pound. If 1 percent $U_3O_8 = 20$ lb U_3O_8 per ton of ore, then 0.1 percent = 2 lb or about \$40.00 to \$86.00 per ton of ore. Lower grade ore may range down to 0.03 percent if 100 tons or more of U_3O_8 can be extracted.

Most of the Oklahoma occurrences are noncommercial except for some of the Permian red-bed deposits, such as that at Cement, Caddo County, in sec. 3, T. 5 N., R. 9 W., where 13 tons of 2.2 percent U_3O_8 was mined from a vein in the Rush Springs Sandstone and shipped to Grants, New Mexico, by the Lister families of Oklahoma City and Chickasha (map no. 46). This ore was commercial at \$6.00/pound only because the U.S. Atomic Energy Commission paid the shipping charges. Many prospects have been examined since the 1950's, and these are listed by counties as follows.

Caddo County.—The Cement deposit is mentioned above. D. C. Shapard, W. H. McConnell, and Jim Hardy, 2307 Classen Boulevard, Oklahoma City, bulldozed a pit and drilled 5 holes 1.8 miles east of Cement in the Rush Springs Sandstone (map no. 47).

Cimarron County.—American Nuclear Corp. of Casper, Wyoming, leased 11,000 acres southwest of Boise City in 1975, supposedly for exploration of the subsurface Triassic red beds. A decision to mine was supposed to have been made in February 1976, according to an article in the *Tulsa World* dated September 14, 1975, but nothing has been mined to date (map no. 9).

Helium is produced from the Keyes sandstone (Morrowan) from 4,630 to 4,873 feet at the Keyes Dome in T. 5 N., R. 8 E.C.M. (map nos. 319–326). The helium composes 2.1 to 2.3 percent of the natural gas and is stored at Cliffside, west of Amarillo, Texas, by the federal government. At present there is a surplus of helium, and the government has curtailed purchases.

Comanche County.—M. Mathis and W. Oberlander of Frederick, Oklahoma, worked a prospect in the Garber Sandstone (Permian) in sec. 31, T. 1 N., R. 15 W., that assayed 120 ppm uranium (map no. 78).

L. C. Mundy, 2109 Yale Street, Wichita Falls, Texas, drilled a 40-foot test on the Kinder Ranch in the S½ sec. 34, T. 1 N., R. 15 W., before 1955 (map no. 64).

Cotton County.—N. B. Nowels, W. E. Handley, H. F. Scheele, and Claude V. Thompson of Ada, Oklahoma, operated the Clinton Byars or Eastman prospect in the SW¼ sec. 30, T. 5 S., R. 12 W., in the Permian Garber Sandstone (map no. 76). The sandstone lens is 25 feet thick by 300 feet wide by 600 feet long. The lower 10 feet contained autunite, bayleyite, carnotite, torbernite, and uranophane; associated minerals were pyrite, chalcopyrite, galena, azurite, and malachite. The samples assayed from 90 to 15,300 ppm uranium.

H. M. Sutterfield, 2133 Avenue I, Wichita Falls, Texas, and O. S. Wilson, 309 Avenue D, Burkburnett, Texas, worked a prospect in sandstone of the Wellington Formation (Permian) on the H. E. Crow Farm, in the SW¹/₄ sec. 7, T. 4 S., R. 12 W., with an

assay of 620 ppm uranium (map no. 99).

Three underground shafts were dug in Wellington sandstone on the Grady Benson Farm in sec. 3, T. 5 S., R. 11 W., 1 mile south of Taylor (map no. 113).

Custer County.—The Red Rock Uranium Co. of Foss mined 20 tons of ore from the basal Doxey silt-stone (Permian) north of Foss in 1955, with assays ranging from 60 to 16,100 ppm uranium (map no. 15). The price received at Grants, New Mexico, was about equal to the cost of shipment.

Johnston County.—J. W. Angel of Wapanucka had a prospect in carbonized wood in sandstone of the Antlers Formation (Cretaceous) in sec. 34, T. 3 S., R. 8 E., with samples assayed at 7,670 ppm uranium (map no. 4).

Nowata County.—In May 1956, the American States Oil Co., southeast of Nowata, invested \$140,000 in a pilot plant to extract uranium and thorium from oil-field waters, but the plant did not go into production, according to Curtis (1956, p. 110).

Pawnee County.—Lee Uto of Galesburg, Kansas, dug 3 pits in sandstone of the Wreford Formation (Pennsylvanian, Gearyan) near the C sec. 8, T. 22 N., R. 4 E., with two 1-foot zones assaying from 800 to 10,700 ppm uranium (map no. 142). The prospect may extend over 100 acres or more.

A few small pits were dug in Wreford sandstone in the $SE^{1/4}$ sec. 18, T. 22 N., R. 4 E., on the W. H. Schone

Farm (map no. 143).

Payne County.—R. B. French of Glencoe dug a 35foot adit in sandstone of the Fort Riley Formation (Pennsylvanian, Gearyan) in the NW1/4 sec. 23, T. 20 N., R. 3 E. (map no. 139).

Roger Mills County.—The Western Oklahoma Uranium Partnership, 3801 Classen Boulevard, Oklahoma City, operated a prospect in the Doxey Shale (Permian) in the $SW^{1/4}NE^{1/4}$ sec. 13, T. 13 N., R. $25 \,\mathrm{W}$, with assays of 70 to 440 ppm uranium (map no.

Rogers County.—The radium springs at Claremore have been used since the early 1900's. The water is from wells 1,075 to 1,110 feet deep in the Devonian Chattanooga Shale (map no. 263).

Tillman County — M. Mathis and W. Oberlander, of Frederick, Oklahoma, dug some surface pits in the Garber Sandstone (Permian) in sec. 1, T. 1 S., R. 15 W., with assays from 600 to $1{,}100$ ppm uranium (map no. 75). They also worked a Garber prospect in sec. 34, T. 2 S., R. 15 W. (map no. 80).

A. H. Messersmith, Box 3133, Monroe Street, Wichita Falls, Texas, dug two pits in the Garber Sandstone on the Barnett Farm in the SE1/4SE1/4 sec.

1, T. 1 S., R. 16 W. (map no. 74).

In 1978, several companies operated prospects in secs. 6, 8, and 16, T. 1 S., R. 15 W., and secs. 12, 13, and 16, T. 1 S., R. 16 W., with F. H. Marbuck of Casper, Wyoming, being one of the lessees (map no. 77). Lloyd Patton, a broker in Chattanooga, stated that four companies, paying \$1.00 to \$5.00 per acre, were drilling tests 1,000 to 1,700 feet deep in northeastern Tillman, southwestern Comanche, and Kiowa Counties, according to articles in The Daily Oklahoman dated May 8 and February 13, 1978.

Tulsa County.—The Third Avenue Uranium Co. of Tulsa tried to mine phosphate nodules from the Coffeyville Formation (Pennsylvanian, Missourian) along the Arkansas River in the 1950's, with assays ranging from 40 to 220 ppm uranium (map no. 169).

Washita County.—The Western Oklahoma Uranium Partnership, 3801 Classen Boulevard, Oklahoma City, worked a prospect in the Doxey siltstone (Permian) in the $NE^{1/4}$ sec. 10, T. 11 N., R. 17 W., with assays ranging from 220 to 420 ppm uranium (map no. 38).

Annotated References

(Numbers are used for cross-reference in tabulation on accompanying large-scale map)

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ranges from 40 to 100 ppm.)

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- 11. Bell, K. G., 1960, Uranium and other trace elements in petroleums and rock asphalts: U.S. Geological Survey Professional Paper 356-B, p. 45-65. (For Oklahoma, 37 crude-oil samples from Ordovician through Permian rocks were analyzed spectrographically, and contained 0.1 to 248.4 ppb uranium. The 45 rock asphalts, ranging from Ordovician through Cretaceous, contained 0.004 to 17.1 ppm uranium in the oil portion, which ranged from 0.58 to 12.28 percent. Several dozen other trace elements are listed. Petroleum may help to concentrate uranium, but it is not a primary source for uranium. The uranium may be attached to the asphaltenes or to colloidal organic material in the oil. Natural paraffin oils contain 1 ppb or less of uranium. The bituminous constituents of rock asphalts average 1,000 ppb uranium. The heavier the gravity of the oil, the greater the uranium content. Hydrogen sulphide is abundant in crude oils and is an excellent agent for reduction. Sea water contains 3 times more uranium than paraffin-based oil.)
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- 27. Campbell, J. A., and Steele, B. A., 1976, Uranium potential of Permian rocks in the southwestern United States: U.S. Geological Survey Open-File Report 76-529, 26 p. (A brief assessment of uranium potential of Permian rocks of the Anadarko Basin of central and western Oklahoma, including a table of occurrences, but excluding chemical data.)
- 28. Chase, G. W., 1954, Occurrence of radioactive material in

sandstone lenses of southwestern Oklahoma: Oklahoma Geological Survey Mineral Report 26, 8 p. (Association of locally abundant radioactive minerals in bituminous gray sandstones of southern Oklahoma. The sandstones belong to the Garber and Wellington Formations, and the minerals are concentrated in the bituminous material.)

29. Curtis, Neville, 1956, Some facts about Oklahoma uranium: Oklahoma Geology Notes, v. 16, p. 106–120. (A summary of knowledge of uranium in Oklahoma and a description of several important occurrences, including the asphaltic deposits south of Sulphur, Murray County. Also includes a reprint of Beroni, 1956.)

30. Daily Oklahoman, 1978, Uranium leases sold: Oklahoma

City, Oklahoma, February 13, p. 30.

31. Daily Oklahoman, 1978, State uranium hunt pressed: Okla-

homa City, Oklahoma, May 8, p. 1-2.

- 32. Danilchik, Walter, and Hyden, H. J., 1957, Pennsylvanian sediments of the Midcontinent region, in Geologic investigations of radioactive deposits, semiannual progress report, December 1, 1956, to May 31, 1957: U.S. Geological Survey Trace Element Investigations Report 690, p. 427–428, issued by U.S. Atomic Energy Commission, Technical Information Service, Oak Ridge, Tennessee. (Pennsylvanian black shales in Oklahoma average 0.001 to 0.002 percent uranium for carbonaceous shales, and 0.002 to 0.009 percent for bituminous shales. Phosphatic nodules in the shales range from 0.009 to 0.023 percent and seem to concentrate the uranium.)
- 33. Donovan, T. J., 1974, Petroleum microseepage at Cement, Oklahoma: evidence and mechanism: American Association of Petroleum Geologists Bulletin, v. 58, p. 429-446. (Contains a brief description of the Permian red beds in the Cement area, Caddo County, showing that the calcite cement is related to petroleum microseepage. An excellent background article for the Cement uranium prospect.)
- 34. Donovan, T. J., Friedman, Irving, and Gleason, J. D., 1974, Recognition of petroleum-bearing traps by unusual isotopic compositions of carbonate-cemented surface rocks: Geology, v. 2, p. 351-354. (Stable isotopic fractionation values might indicate vertical migration and subsequent oxidation of low-molecular-weight hydrocarbons. The carbon isotope values for the Cement Oil Field, Caddo County, range from -5.5 to -39.2 permil PDB. These light values suggest that hydrocarbons seeped slowly upward.)

 Duhme, R. M., and Meek, W. B., 1956, Preliminary report on uranium occurrences: U.S. Geological Survey Prelimi-

nary Reconnaissance Report DAO-P-6, 2 p.

- 36. Eargle, D. H., and McKay, E. J., 1955, Permian and Triassic sediments of northern Texas and southern Oklahoma, in Geologic investigations of radioactive deposits, semiannual progress report, June 1 to November 30, 1955: U.S. Geological Survey Trace Element Investigations Report 590, p. 257–262, issued by U.S. Atomic Energy Commission, Technical Information Service, Oak Ridge, Tennessee. (In southwestern Oklahoma, three stratigraphic units are uraniferous: (1) the Wichita or Garber and Wellington Formations include channel sandstones with uranium and copper minerals; (2) the Hennessey Group has uraniferous bituminous nodules in shale; and (3) the upper Cloud Chief Formation south of Clinton, Custer County, has uranium minerals in thin sandstone beds.)
- 37. Erickson, R. L., Myers, A. T., and Horr, C. A, 1954, Association of uranium and other metals with crude oil, asphalt, and petroliferous rock: American Association of Petroleum Geologists Bulletin, v. 38, p. 2200–2218. (Chemical analyses of crude oils, asphalts, and petroliferous rocks show that the uranium in crude oils is much less than that of natural asphalts and heavy oil extracted from petroliferous rocks. Trace metals, such as vanadium, are consistently concentrated in organic matter in natural asphalts and heavy extracted oils. The Page grahamite deposit, in southern Le Flore County, has 0.7 ppm uranium.)

38. Fay, R. O., and Hart, D. L., Jr, 1978 [1979], Geology and mineral resources (exclusive of petroleum) of Custer County, Oklahoma: Oklahoma Geological Survey Bulletin 114, 88 p. (Gives uranium and vanadium analyses of basal Doxey

siltstone in the Foss-Hammon area.)

Ferguson, J. D., 1979, The subsurface alteration and mineralization of Permian red beds overlying several oil fields in southern Oklahoma: Shale Shaker, v. 29, pt. 1, p. 172–178; v. 29, pt. 2, p. 200–208. Also Oklahoma State University M.S.

thesis, 96 p., 1977. (Pyrite and calcite cement may form over oil fields, perhaps owing to reactions as follows: $C_nH_m + 3CaSO_4 \rightarrow 3CaCO_3 + CO_2 + H_2O + 3H_2S$ and $2H_2S + Fe_2O_3 \rightarrow FeS_2 + FeO + 2H_2O$. Of interest is that oil may migrate as droplets on water.)

40. Finch, W. I., 1956, Uranium in terrestrial sedimentary rocks in the United States exclusive of the Colorado Plateau, in Page, L. R., Stocking, H. E., and Smith, H. B., compilers, Contributions to the geology of uranium and thorium by the United States Geological Survey and Atomic Energy Commission for the United Nations International Conference on Peaceful Uses of Atomic Energy, Geneva, Switzerland, 1955: U.S. Geological Survey Professional Paper 300, p. 321-327 (1955). (Descriptions of occurrences of uranium in the Permian Garber Sandstone in Tillman, Cotton, and Jefferson Counties, Oklahoma. The uranium deposits are of two types: (1) copper and uranium minerals in fine-grained sandstones and (2) uranium associated with asphaltic arkosic sandstones. The uraniferous zones are almost always in light-colored lenses. The main minerals are torbernite, autunite, uranophane, malachite, and azurite.)

41. Finch, W. I., 1967, Geology of epigenetic uranium deposits in sandstone in the United States: U.S. Geological Survey Professional Paper 538, 121 p. (This study includes discussions on lithology and depositional history of host rocks, character of the deposits, and origins. Occurrences reported in Beck-

ham, Caddo, and Washita Counties, Oklahoma.)

42. Fischer, R. P., and Stewart, J. H., 1960, Distribution and lithologic characteristics of sandstone beds that contain deposits of copper, vanadium, and uranium, in Geological Survey Research, 1960: U.S. Geological Survey Professional Paper 400-B, p. 42-44. (Oklahoma red-bed occurrences classed as copper and copper-uranium stream deposits. These are first-cycle sediments derived from a granitic rock source. Some rocks have a trace of vanadium. The copper and uranium were probably precipitated from circulating

ground waters by reducing agents.)

43. Garrels, R. M., Hostetler, P., and Christ, C. L., 1957, The system U-H₂O-CO₂, in Geologic investigations of radioactive deposits, semiannual progress report, December 1, 1956, to May 31, 1957: U.S. Geological Survey Trace Element Investigations Report 690, p. 550, issued by U.S. Atomic Energy Commission, Technical Information Service, Oak Ridge, Tennessee. (A portion of the pH-Eh-P_{CO2} diagram of the U-H₂O-CO₂ system was constructed from thermodynamic data and is shown in figure 115. The information confirms previous reports (TEI-620, p. 319) that uranium minerals may have been deposited from uranyl dicarbonate complexes by reduction with carbonaceous material. This article is quoted by some authors to explain the possible origin for uranium in many Oklahoma deposits.)

44. Gott, G. B., Wyant, D. G., and Beroni, E. P., 1952, Uranium in black shales, lignites, and limestones in the United States: U.S. Geological Survey Circular 220, p. 31–35. (The uranium in the Chattanooga black shale of Tennessee and Kentucky ranges between 0.001 and 0.03 percent. The uranium is greater with greater organic material. The gammaray logs of the Chattanooga and Woodford units in Oklahoma are of the same order of radioactivity as that of the

Chattanooga of the eastern United States.)

45. Gott, G. B., and Hill, J. W., 1953, Radioactivity in some oil fields of southeastern Kansas: U.S. Geological Survey Bulletin 988-E, p. 69–122. (Radium-bearing precipitates have been found in oil-well fluids in southern Kansas. The association of helium, probably radiogenic, suggests a high concentration of uranium. Recent leaching of the uranium is possible because the equilibrium of 0.5 percent uranium is never reached. The radioactivity ranges between 0 and 10.85 percent equivalent uranium oxide (%eU₃O₈), with the uranium oxide content ranging between 0 and 0.006 percent.)

46. Hail, W. J., Jr., 1954, Asphaltic rocks in the western states, in Geologic investigations of radioactive deposits, semi-annual progress report, December 1, 1953, to May 31, 1954: U.S. Geological Survey Trace Element Investigations Report 440, p. 142–143, issued by U.S. Atomic Energy Commission, Technical Information Service, Oak Ridge, Tennessee. (An occurrence of Garber uraniferous asphaltic sandstone near Asphaltum, Jefferson County, Oklahoma. A grab sample had 0.0125 percent cU in 0.97 percent ash.)

47. Hail, W. J., Jr., 1955, Asphaltic rocks in the western states,

in Geologic investigations of radioactive deposits, semiannual progress report, December 1, 1954, to May 31, 1955: U.S. Geological Survey Trace Element Investigations Report 540, p. 175–177, issued by U.S. Atomic Energy Commission, Technical Information Service, Oak Ridge, Tennessee. (The ash of the extracted oil from asphalt-bearing rocks of the Sulphur area, Murray County, contained 0.22 percent uranium.)

48. Hail, W. J., Jr., Myers, A. T., and Horr, C. A., 1956, Uranium in asphalt-bearing rocks of the western United States, in Page, L. R., Stocking, H. E., and Smith, H. B., compilers, Contributions to the geology of uranium and thorium by the United States Geological Survey and Atomic Energy Commission for the United Nations International Conference on Peaceful Uses of Atomic Energy, Geneva, Switzerland, 1955: U.S. Geological Survey Professional Paper 300, p. 521-526 (1955). (Uranium is concentrated as organouranium complexes in the asphalt and not in the host rock. The amount of uranium in the asphalt is probably related to the composition of the source sediments of the asphalts, or to those rocks through which the oil migrated. Oklahoma uraniferous asphalt-bearing rocks are largely sandstones. Samples from the Sulphur area, Murray County, from ash of the extracted oil from the asphaltic rock, ranged from 0.001 to

49. Hail, W. J., Jr., 1959, Reconnaissance for uranium in asphalt-bearing rocks in the western United States: U.S. Geological Survey Bulletin 1046, p. 55-83. (Contains brief descriptions of Sulphur and Cameron deposits, and 15 other less significant deposits, and includes a table of chemical

analyses.)

 Haines, G. I., 1955, Preliminary report on uranium occurrences: U.S. Geological Survey Preliminary Reconnaissance

Report DEB-P-5-2426, 2 p.

51. Hanson, R. E., 1977, Petrology and geochemistry of the Carlton Rhyolite, southern Oklahoma: Oklahoma State University unpublished M.S. thesis, 63 p. (Sixty-eight samples were collected from the Arbuckle and Wichita Mountains. The uranium averaged 3 ppm, ranging from 2 to 16 ppm. The rhyolites and granites of the Wichita province are suggested as possible source rocks for uranium mineralization in adjacent sedimentary rocks.)

- 52. Hill, J. W., 1953, South-central district, in Search for and geology of radioactive deposits, semiannual progress report, December 1, 1952, to May 31, 1953: U.S. Geological Survey Trace Element Investigations Report 330, p. 200–204, issued by U.S. Atomic Energy Commission, Technical Information Service, Oak Ridge, Tennessee. (Tar seeps from the Pennsylvanian Cherokee Group were found to contain 0.04 percent cU in the 0.73 percent ash in the Picher Field, Ottawa County. Unweathered asphaltic pellets from Permian red beds near the Wichita Mountains contain up to 9.38 percent cU in the 9.20 percent ash. The original source for the uranium might be related to the nearby igneous rocks. In Pawnee County, selected samples from lignitic lenses in cupriferous Permian sandstones contain 16.3 percent cU [this seems too high].)
- Hill, J. W., and Kaiser, E. P., 1953, Preliminary report on uranium occurrences: U.S. Geological Survey Preliminary Reconnaissance Report D-748, 2 p.

 Hill, J. W., and Rose, Arthur, 1953, Preliminary report on uranium occurrences: U.S. Geological Survey Preliminary

Reconnaissance Report D-744, 2 p.

55. Hill, J. W, 1954, Uraniferous asphaltic materials of southwestern Oklahoma [abstract]: Geological Society of America Bulletin, v. 65, p. 1377. (Uraniferous asphaltic pellets in Permian arkosic red-brown shales and sandstones are described. Many pellets contain smaltite and uraninite, with uranium and total radioactivity evenly distributed within the pellet. The close correlation between asphaltic pellets and asphalt seeps suggests that the pellets are derived from such a source.)

56. Hill, J. W., 1957, Uranium-bearing carbonaceous nodules of southwestern Oklahoma: Oklahoma Geological Survey Mineral Report 33, 6 p. (Permian rocks on the north flank of the Wichita Mountains contain uraniferous carbonaceous nodules. The uranium was uniformly distributed in the nodules, with amounts ranging from 0.37 to 0.86 percent cU. The trace-metal content of the nodules is similar to that of crude oils in the area.)

57. Huang, W. T., 1956, Novacekite from the Wichita Mountains, Oklahoma: American Mineralogist, v. 41, p. 152–153.

58. Hyden, H. J., and Bass, N. W., 1954, Uranium in western petroleum, in Geologic investigations of radioactive deposits, semiannual progress report, June 1 to November 30, 1954: U.S. Geological Survey Trace Element Investigations Report 490, p. 185–187, issued by U.S. Atomic Energy Commission, Technical Information Service, Oak Ridge, Tennessee. (Four samples of oil field asphalts from Oklahoma ranged from 0.014 percent ash with 0.0003 percent uranium to 0.058 percent ash with 0.001 percent uranium.)

59. Hyden, H. J., and Woods, N. W., 1955, Uranium in petroleum in the western United States, in Geologic investigations of radioactive deposits, semiannual progress report, December 1, 1954, to May 31, 1955; U.S. Geological Survey Trace Element Investigations Report 540, p. 179–183, issued by U.S. Atomic Energy Commission, Technical Information Service, Oak Ridge, Tennessee. (The ash of crude oil from the Bartlesville sand of the Nowata Oil Field averaged 400 to 450 ppm uranium. The average uranium from ash of crude oils in sandstone reservoirs in Oklahoma is 81 ppm. Oil field brines from Nowata range from 6 ppb to less than 1 ppb. Based on the lack of appreciable amounts of nitrogen in oil (0.018 to 0.48 percent) compared to uranium in ash (1-75 ppm), the uranium is not expected to exist as

uranium-porphyrin complexes in the oil.)

60. Hyden, H. J., and Danilchik, Walter, 1955, Midcontinent Pennsylvanian shales, in Geologic investigations of radioactive deposits, semiannual progress report, June 1 to November 30, 1955: U.S. Geological Survey Trace Element Investigations Report 590, p. 253–257, issued by U.S. Atomic Energy Commission, Technical Information Service, Oak Ridge, Tennessee. (Pennsylvanian rocks in northeastern Oklahoma were sampled for radioactivity. Bituminous black shales with phosphatic nodules were the most persistent highly radioactive rocks. The Excello Shale of the upper Cherokee Group ranged from 0.0005 to 0.01 percent uranium. Phosphatic nodules in the Excello ranged up to 0.025 percent uranium.)

61. Hyden, H. J., 1956, Uranium and other trace metals in crude oils of the western United States, in Page, L. R., Stocking, H. E., and Smith, H. B., compilers, Contributions to the geology of uranium and thorium by the United States Geological Survey and Atomic Energy Commission for the United Nations International Conference on Peaceful Uses of Atomic Energy, Geneva, Switzerland, 1955: U.S. Geological Survey Professional Paper 300, p. 511–519 (1955). (Crude-oil samples from different-age reservoirs in north-central Oklahoma were fluorimetrically analyzed for their uranium content. The uranium in oil is not preferentially distributed with respect to age, lithology, rock type, or type of oil. Vanadium and nickel are present as porphyrin complexes in oil, but uranium is not.)

 Hyden, H. J., and Danilchik, Walter, 1962, Uranium in some rocks of Pennsylvanian age in Oklahoma, Kansas, and Missouri: U.S. Geological Survey Bulletin 1147-B, p. 1–82.

- 63. Landis, E. R., 1954, Western Kansas and eastern Colorado, in Geologic investigations of radioactive deposits, semi-annual progress report, June 1 to November 30, 1954: U.S. Geological Survey Trace Element Investigations Report 490, p. 158–159, issued by U.S. Atomic Energy Commission, Technical Information Service, Oak Ridge, Tennessee. (Black shales in the lower Pierre Formation of western Kansas and eastern Colorado contain not more than 0.008 percent uranium. Tuffaceous and carbonate rocks were also analyzed. A silicified limestone from the Ogallala Formation in Clark County, Kansas, contained 0.01 percent uranium.)
- 54. Landis, E. R., 1955, Midcontinent Devonian shale, in Geologic investigations of radioactive deposits, semiannual progress report, June 1 to November 30, 1955: U.S. Geological Survey Trace Element Investigations Report 590, p. 249—252, issued by U.S. Atomic Energy Commission, Technical Information Service, Oak Ridge, Tennessee. (Gamma-ray logs were used to evaluate the Devonian-Mississippian Chattanooga black shale in Kansas. Equivalent uranium values ranged from 0.004 to 0.014 percent eU. Radioactivity is greatest in the lowermost part of the unit and increases in a southerly direction toward Oklahoma.)

65. Landis, E. R., 1957, Uranium content of ground and surface

waters in western Kansas, eastern Colorado, northeastern New Mexico, and the Oklahoma Panhandle, in Geologic investigations of radioactive deposits, semiannual progress report, December 1, 1956, to May 31, 1957: U.S. Geological Survey Trace Element Investigations Report 690, p. 539-542, table 37, issued by U.S. Atomic Energy Commission, Technical Information Service, Oak Ridge, Tennessee. (Same as 1961 article.)

66. Landis, E. R., 1961, Uranium content of ground and surface waters in a part of the central Great Plains, in Contributions to the geology of uranium, 1960: U.S. Geological Survey Bulletin 1087-G, p. 223-258. (Different waters from different rocks have different average uranium values. Waters in Cenozoic tuffaceous fluviatile rocks average 6.7 ppb uranium; in Cretaceous shales the waters average 20.4 ppb uranium; and in terrestrial and nearshore sandstones and siltstones the waters average 10.2 ppb uranium. Several water samples from the Oklahoma Panhandle range from 1 to 41 ppb uranium.)

67. Landis, E. R., 1962, Uranium and other trace elements in Devonian and Mississippian black shales in the central Midcontinent area: U.S. Geological Survey Bulletin 1107-E, p.

68. Lane Wells Company, sample log-9/14/15; gamma-ray log-10/27/54.

69. Lockwood, R. P., 1976 [1977], Geochemistry and petrology of some Oklahoma red-bed copper occurrences, in Johnson, K. S., and Croy, Rosemary, editors, Stratiform copper deposits of the Midcontinent region, a symposium: Oklahoma Geological Survey Circular 77, p. 61–68. (Two copper-shale samples from the Permian red beds around the Wichita Mountains ranged between 0.007 and 0.01 weight percent uranium.)

70. Moxham, R. M., 1953, Airborne radioactivity surveying, in Search for and geology of radioactive deposits, semiannual progress report, December 1, 1952, to May 31, 1953: U.S. Geological Survey Trace Element Investigations Report 330, p. 280-292, issued by U.S. Atomic Energy Commission, Technical Information Service, Oak Ridge, Tennessee. (Mentions plans for an airborne gamma-ray map of the

Wichita Mountains area.)

71. McKay, E. J., and Hyden, H. J., 1956, Permian of north Texas and southern Oklahoma, in Geologic investigations of radioactive deposits, semiannual progress report, June 1 to November 30, 1956: U.S. Geological Survey Trace Element Investigations Report 640, p. 208-216, issued by U.S. Atomic Energy Commission, Technical Information Service, Oak Ridge, Tennessee. (A brief description of the lithology and structure of the Permian Rush Springs sandstones in the Cement Field, Caddo County, is given, relating mineral types to color variations. Radioactivity along a mined trench on the Cement Dome ranges from 0.05 to 0.8 mr/hr.)

72. McKay, E. J., 1957, Reconnaissance for uranium in northern Texas and southern Oklahoma: U.S. Geological Survey Trace Element Investigations Report 689, 31 p., issued by U.S. Atomic Energy Commission, Technical Information Service, Oak Ridge, Tennessee. (A description of the uranium potential of southern Oklahoma, including the Cement Field, is given, with chapters on stratigraphy and regional structure of the area. The Byars deposit, in the Garber Sandstone of Cotton County, and the Cement deposit, in the Rush Springs Formation of Caddo County, are described, including mineralogy, radioactivity, and size of prospects. The Byars prospect ranges from 0.001 to 0.008 percent uranium.)

73. McKay, E. J., 1957, Permian of north Texas and southern Oklahoma, in Geologic investigations of radioactive deposits, semiannual progress report, December 1, 1956, to May 31, 1957: U.S. Geological Survey Trace Element Investigations Report 690, p. 425-427, issued by U.S. Atomic Energy Commission, Technical Information Service, Oak Ridge, Tennessee. (Two Permian uraniferous sandstone deposits are described, one in the Garber Sandstone of the Red River area, and one in the Rush Springs Formation at Cement, Caddo County. A red-bed facies favors the concentration of uranium.)

74. McKelvey, V. E., and Nelson, J. M., 1950, Characteristics of marine uranium-bearing sedimentary rocks: Economic Geology, v. 45, p. 35-53. (Marine black shales have 0.01 to 0.02 percent uranium, especially those rich in sulfides and

organic matter, but low in carbonate. In general, uranium increases with an increase in phosphate, but the nature of the uranium mineral in the phosphate is unknown. The Woodford Shale of Oklahoma is an analogous unit.)

75. McKelvey, V. E., Everhart, D. L., and Garrels, R. M., 1956, Summary of hypotheses of genesis of uranium deposits, in Page, L. R., Stocking, H. E., and Smith, H. B., compilers, Contributions to the geology of uranium and thorium by the United States Geological Survey and Atomic Energy Commission for the United Nations International Conference on Peaceful Uses of Atomic Energy, Geneva, Switzerland, 1955: U.S. Geological Survey Professional Paper 300, p. 41-53 (1955). (Sedimentary uranium deposits generally contain more vanadium than igneous rocks. The precipitation of uranium is likely due to reduction related to decaying organic matter. The ultimate source of the uranium may be devitrified volcanic ash, weathered igneous rocks, or hydrothermal solutions. The uranium is probably adsorbed by humates and other organic compounds from solutions migrating through them. The precipitation in phosphorites takes place by chemical adsorption by apatite. Reference is made to the Cherokee shale of Oklahoma.)

76. Mickle, D. G., editor, 1978, Classification of uranium deposits: Bendix Field Engineering Corp., Grand Junction, Colorado, 77 p. (A summary is given of uranium occurrences in sedimentary, plutonic, and volcanic rocks, and in deposits of uncertain genesis. These main groups are subdivided into subclasses, including expected uranium content for most

types of deposits.)

77. Morrison, C. M., 1980, Permian uranium-bearing sandstones on the Muenster-Waurika Arch and in the Red River

area: Shale Shaker, v. 30, p. 143-156.

78. Olmsted, R. W., 1975, Geochemical studies of uranium in south-central Oklahoma: Oklahoma State University unpublished M.S. thesis, 116 p. (Uraniferous Permian sandstones are described in the Cement and Cox City areas of Caddo and Grady Counties. The geochemical aspects of ore genesis in relationship to hydrocarbon migration were studied, especially by analyzing ground-water samples. The original source for the uranium was probably igneous rocks eroded from the Wichita and Arbuckle Mountains. Some ground-water samples at Cement had as much as 800 ppb uranium.)

79. Patrick, D. M., and Ham, W. E., 1969, Oklahoma, in Uranium in the southern United States: Southern Interstate Nuclear Board and U.S. Atomic Energy Commission, p. 36-56. (A summary is given of uranium occurrences in Oklahoma with descriptions of stratigraphic units, including tables of locations, types of occurrences, and percent uranium. The

uranium potential of the State is discussed.)

80. Pierce, A. P., 1954, Radon and helium studies, in Geologic investigations of radioactive deposits, semiannual progress report, June 1 to November 30, 1954: U.S. Geological Survey Trace Element Investigations Report 490, p. 274-276, issued by U.S. Atomic Energy Commission, Technical Information Service, Oak Ridge, Tennessee. (The genesis and trace-metal content of asphaltites in Pennsylvanian and Permian rocks along the Amarillo-Wichita Uplift are discussed. Most of the trace metals occur as inclusions of arsenide and sulphide minerals. The inclusions were apparently formed before solidification of the asphaltite, which would be evidence that the asphalt passed through a liquid phase.)

81. Pierce, A. P., Mytton, J. W., and Gott, G. B., 1956, Radioactive elements and their daughter products in the Texas Panhandle and other oil and gas fields in the United States, in Page, L. R., Stocking, H. E., and Smith, H. B., compilers, Contributions to the geology of uranium and thorium by the United States Geological Survey and Atomic Energy Commission for the United Nations International Conference on Peaceful Uses of Atomic Energy, Geneva, Switzerland, 1955: U.S. Geological Survey Professional Paper 300, p. 527-532 (1955). (The occurrence of radon, helium, and radium in uraniferous asphaltic rocks along the Amarillo-Wichita Uplift is discussed. The radium in oil-field waters is as great as 1×10^{-9} grams of radium per liter. The uranium content in oil-field brines is as much as 0.2 ppm.)

82. Pierce, A. P., Gott, G. B., and Mytton, J. W., 1964, Uranium and helium in the Panhandle Gas Field, Texas and adjacent areas: U.S. Geological Survey Professional Paper

454-G, 57 p.

82a. Rodman, M. R., Gordon, L. I., and Chen, A. C. T., 1979, Extraction of uranium from sea water, *in* Evaluation of uranium resources and plant siting: U.S. Department of

Energy, v. 1, Grand Junction, Colorado.

83. Rogers, A.S., 1953, Physical behavior of radon, in Search for and geology of radioactive deposits, semiannual progress report, December 1, 1952, to May 31, 1953: U.S. Geological Survey Trace Element Investigations Report 330, p. 293—296, issued by U.S. Atomic Energy Commission, Technical Information Service, Oak Ridge, Tennessee. (The phase relationships between radon and water and different gases were studied, giving the radon and radium content of brine samples from several wells in Texas County, Oklahoma.)

 Rose, Arthur, 1953, Preliminary report on uranium occurrences: U.S. Geological Survey Preliminary Reconnaissance

Report D-745, 2 p.

 Russell, R. T., 1954, Preliminary report on uranium occurrences: U.S. Geological Survey Preliminary Reconnaissance Report DEB-C-1640, 2 p.

 Russell, R. T., 1955, Preliminary report on uranium occurrences: U.S. Geological Survey Preliminary Reconnaissance

Report DEB-C-1652, 2 p.

- 87. Russell, W. L., 1945, Relation of radioactivity, organic content, and sedimentation: American Association of Petroleum Geologists Bulletin, v. 29, p. 1470–1493. (A relationship exists between radioactivity and types of organic content. Marine oil shales generally have high radioactivity, coals abnormally low radioactivity, and other types of organic matter intermediate radioactivity. There is no evidence for an increase in radioactivity with slowness of deposition.)
- 88. Scott, R. C., and Barker, F. B., 1962, Data on uranium and radium in ground water in the United States, 1954 to 1957: U.S. Geological Survey Professional Paper 426, 115 p. (The uranium in ground water from a sample from the Rush Springs sandstone at Hinton, Caddo County, was 7.4 micrograms per liter.)

 Siebenthal, C. E., 1908, Mineral resources of northeastern Oklahoma: U.S. Geological Survey Bulletin 340, p. 187–228.
 (Locations and depths of the radium-springs water wells at Claremore, Rogers County, are given, p. 226–227.)

- 90. Slaughter, A. L., and Clabaugh, S. E., 1945, Preliminary report on a trace elements reconnaissance in central and southwestern states: U.S. Geological Survey Trace Element Investigations Report 9, 59 p., issued by U.S. Atomic Energy Commission, Technical Information Service, Oak Ridge, Tennessee.
- Stafford, H. S., and Beroni, E. P., 1956, Preliminary report on uranium occurrences: U.S. Geological Survey Preliminary Reconnaissance Report DAO-P-6-2809, 2 p.
- 92. Stafford, H. S., and Meek, W. B., 1956, Preliminary report on uranium occurrences: U.S. Geological Survey Preliminary Reconnaissance Report DAO-P-6-2807 and 2808, 2 p. each.
- 93. Stocking, H. E., and Page, L. R., 1956, Natural occurrence of uranium in the United States—a summary, in Page, L. R., Stocking, H. E., and Smith, H. B., compilers, Contributions to the geology of uranium and thorium by the United States Geological Survey and Atomic Energy Commission for the United Nations International Conference on Peaceful Uses of Atomic Energy, Geneva, Switzerland, 1955: U.S. Geological Survey Professional Paper 300, p. 5–12 (1955). (A summary is given of types and locations of uranium occurrences in the United States, including a brief discussion of uraniferous asphaltites and clastic sandstones in Oklahoma.)
- 94. Swanson, V. E., 1954, Midcontinent Devonian shales, in Geologic investigations of radioactive deposits, semiannual progress report, June 1 to November 30, 1954: U.S. Geological Survey Trace Element Investigations Report 490, p. 160–164, issued by U.S. Atomic Energy Commission, Technical Information Service, Oak Ridge, Tennessee. (Seventy-five samples were collected from the Woodford Shale of

Oklahoma, averaging 0.003 percent uranium.)

95. Swanson, V. E., 1955, Midcontinent Devonian shales, in Geologic investigations of radioactive deposits, semiannual progress report, December 1, 1954, to May 31, 1955: U.S. Geological Survey Trace Element Investigations Report 540, p. 169–170, issued by U.S. Atomic Energy Commission, Technical Information Service, Oak Ridge, Tennessee. (The Chattanooga and Woodford black shales of the Tahlequah—Spavinaw area of northeastern Oklahoma and Arbuckle Mountains of southern Oklahoma were studied, using samples, cores, and gamma-ray logs. No data were released.)

96. Swanson, V. E., 1956, Uranium in marine black shales of the United States, in Page, L. R., Stocking, H. E., and Smith, H. B., compilers, Contributions to the geology of uranium and thorium by the United States Geological Survey and Atomic Energy Commission for the the United Nations International Conference on Peaceful Uses of Atomic Energy, Geneva, Switzerland, 1955: U.S. Geological Survey Professional Paper 300, p. 451–456 (1955). (The uranium potential of marine black shales is dependent upon the tectonic and paleogeographic setting of the depositional environment of the organic muds. The most uraniferous marine shales are those deposited in epicontinental areas such as the Chattanooga and Woodford Shales of Oklahoma. The Woodford averages 0.005 percent uranium, and some samples range up to 0.008 percent uranium.)

97. Swanson, V. E., 1960, Oil yield and uranium content of black shales, in Uranium in carbonaceous rocks: U.S. Geological Survey Professional Paper 356-A, p. 1–44. (The relationship of oil yield to uranium content of Pennsylvanian black shales of northeastern Oklahoma and the Woodford Shale of the Arbuckle Mountains is discussed.)

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