ALTERATION OF SANDSTONE AS A GUIDE TO URANIUM DEPOSITS AND THEIR ORIGIN, NORTHERN BLACK HILLS, SOUTH DAKOTA

By R. C. Vickers

Trace Elements Memorandum Report 568

UNITED STATES DEPARTMENT OF THE INTERIOR

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November 15, 1956

AEC-174/7

Mr. Robert D. Nininger
Assistant Director for Exploration
Division of Raw Materials
U. S. Atomic Energy Commission
Washington 25, D. C.

Dear Bob:

Transmitted herewith are three copies of TEM-568, "Alteration of sandstone as a guide to uranium deposits and their origin, northern Black Hills, South Dakota," by R. C. Vickers, July 1956.

We are asking Mr. Hosted to approve our plan to submit this report for publication in Economic Geology.

Sincerely yours,

John H. Eric

for W. H. Bradley
Chief Geologist

Geology and Mineralogy

This document consists of 24 pages,
plus 1 figure.
Series A

UNITED STATES DEPARTMENT OF THE INTERIOR

GEOLOGICAL SURVEY

ALTERATION OF SANDSTONE AS A GUIDE TO URANIUM DEPOSITS AND THEIR ORIGIN, NORTHERN BLACK HILLS, SOUTH DAKOTA*

By

R. C. Vickers

July 1956

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*This report concerns work done on behalf of the Division of Raw Materials of the U. S. Atomic Energy Commission.

USGS - TEM-568

GEOLOGY AND MINERALOGY

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ALTERATION OF SANDSTONE AS A GUIDE TO URANIUM DEPOSITS AND THEIR ORIGIN, NORTHERN BLACK HILLS, SOUTH DAKOTA

By R. C. Vickers

ABSTRACT

Several uranium deposits are present in the Fall River sandstone of Early Cretaceous age on the northeast flank of the Black Hills, Butte County, South Dakota. The deposits are within a fine-grained, well-sorted, persistent basal sandstone unit that ranges in thickness from 2 to 18 feet and dips about 4° NE.

Detailed mapping of about 2 square miles surrounding the deposits has shown that all the uranium occurrences and most of the areas of high radioactivity are where the color changes in the basal sandstone from reddish on the up-dip side of the occurrences to yellowish-gray or buff down-dip. Radioactivity measurements show that uranium is distributed almost continuously along the sinuous red-buff contact for more than 5 miles. Laboratory work indicates that the red color is caused by hematite resulting from the alteration of ferrous iron minerals and hydrous ferric oxides.

The close association of the red-buff contact and the uranium deposits suggests that the two were formed by the same solutions. The uranium was probably deposited originally from ground water which moved down-dip and gradually changed from an oxidizing solution near the surface to a mildly reducing solution at depth. Concentrations of uranium have resulted from the localization of reducing conditions caused perhaps by structures superimposed on the regional dip, local thinning or decrease in permeability of the sandstone, or concentrations of pyritiferous carbonaceous material.

The red alteration is probably the result of pre-Oligocene weathering that has extended downward in the more permeable beds about 200 feet below the ancient erosion surface.

Oxidation of the primary uranium during the present weathering cycle has resulted in the formation of carnotite and possibly other secondary uranium minerals.

INTRODUCTION

Abnormal radioactivity in southwestern Butte County, South Dakota was first detected in August 1953 by light-plane airborne reconnaissance conducted by the U. S. Atomic Energy Commission. Later, ground reconnaissance by geologists of the Commission and prospectors resulted in the discovery of several small sandstone-type uranium deposits. During the winter of 1953-54, about 450 tons of rock containing between 0.1 and 0.2 percent uranium was mined from one of the deposits (Kling No. 2), and about 8 tons of similar-grade material was mined from the Bonato deposit. There has been no additional production from the area to date.

During August 1954, about 2 square miles surrounding the Kling and Bonato uranium deposits were mapped by the U. S. Geological Survey with plane table and alidade at a scale of 1/2400 to determine ore controls that might be used as guides to find additional deposits.

Location and general features

The area of investigation is shown in figure 1. The locality is on the northeast flank of the Black Hills uplift, about 6 miles west of Belle Fourche, and includes parts of secs. 23, 24, 25, and 26, T. 8 N., R. 1 E., Butte County, South Dakota.

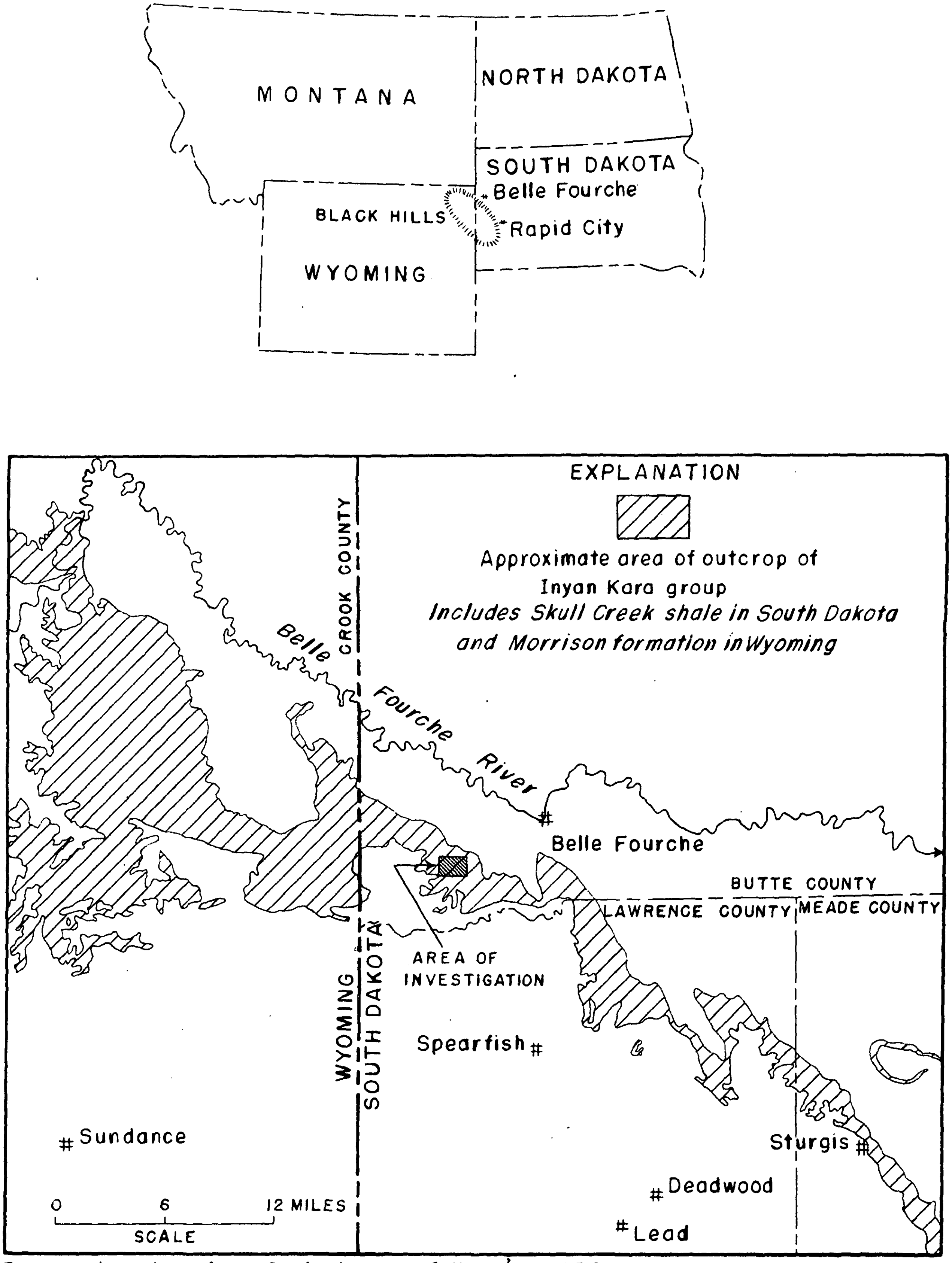
The area has a slightly undulating surface that slopes gently to the northeast and is dissected by numerous small canyons generally less than 150 feet deep. The average surface elevation is about 3,400 feet, and the total relief in the mapped area is about 900 feet.

Acknowledgments

Laboratory space and equipment of the Geology Department of the University of Wisconsin were used for the preparation and study of rock samples. The use of these facilities is greatly appreciated.

The author is grateful for the very able assistance of Pierce D. Parker, U. S. Geological Survey during the field work.

This work was done by the U. S. Geological Survey on behalf of the Division of Raw Materials of the U. S. Atomic Energy Commission.



Base and geology from Geologic map of Wyoming, 1952
and Geologic map of South Dakota, 1951

FIG. 1.-INDEX MAP SHOWING LOCATION OF THE AREA OF INVESTIGATION, BUTTE COUNTY, S. DAK.

GEOLOGY

The general geology is relatively simple. The basal sandstone unit of the Early Cretaceous Fall River sandstone of the Inyan Kara group forms an almost continuous outcrop along the canyon sides and rims. The Fuson shale, which underlies the Fall River formation, crops out in the lower parts of the canyons. The general stratigraphy of the area and a typical section in one of the numerous small canyons are shown in figure 2. The rocks dip about 4° NE and dip-slope surfaces are developed on sandstone beds of the Fall River formation in the intercanyon areas.

Fall River sandstone

The most persistent unit in the Fall River sandstone is the basal sandstone, which ranges from 2 to 18 feet in thickness but generally is from 6 to 10 feet thick, and is composed of fine-grained, massive, generally uniformly-bedded buff to red sandstone. Size analyses show that the sand is well sorted; more than 95 percent of the grains are fine sand (1/4 to 1/8 mm) and very fine sand (1/8 to 1/16 mm); the remainder is silt and clay. Carbonaceous plant fragments are locally abundant in small lenses or along bedding planes. Vertical holes, a few millimeters in diameter, some of which are partly filled with carbonaceous material, were observed in the upper few inches of the basal unit, and these may represent a carbonized root system. A thin bed of shaly lignite, a few inches to 1 foot thick, overlies the sandstone.

The lithology of the Fall River sandstone above the shaly lignite consists mainly of variable amounts of impure sandstones, siltstones, and shales. About 1 mile southeast of the Bonato ranch (fig. 3), the upper part

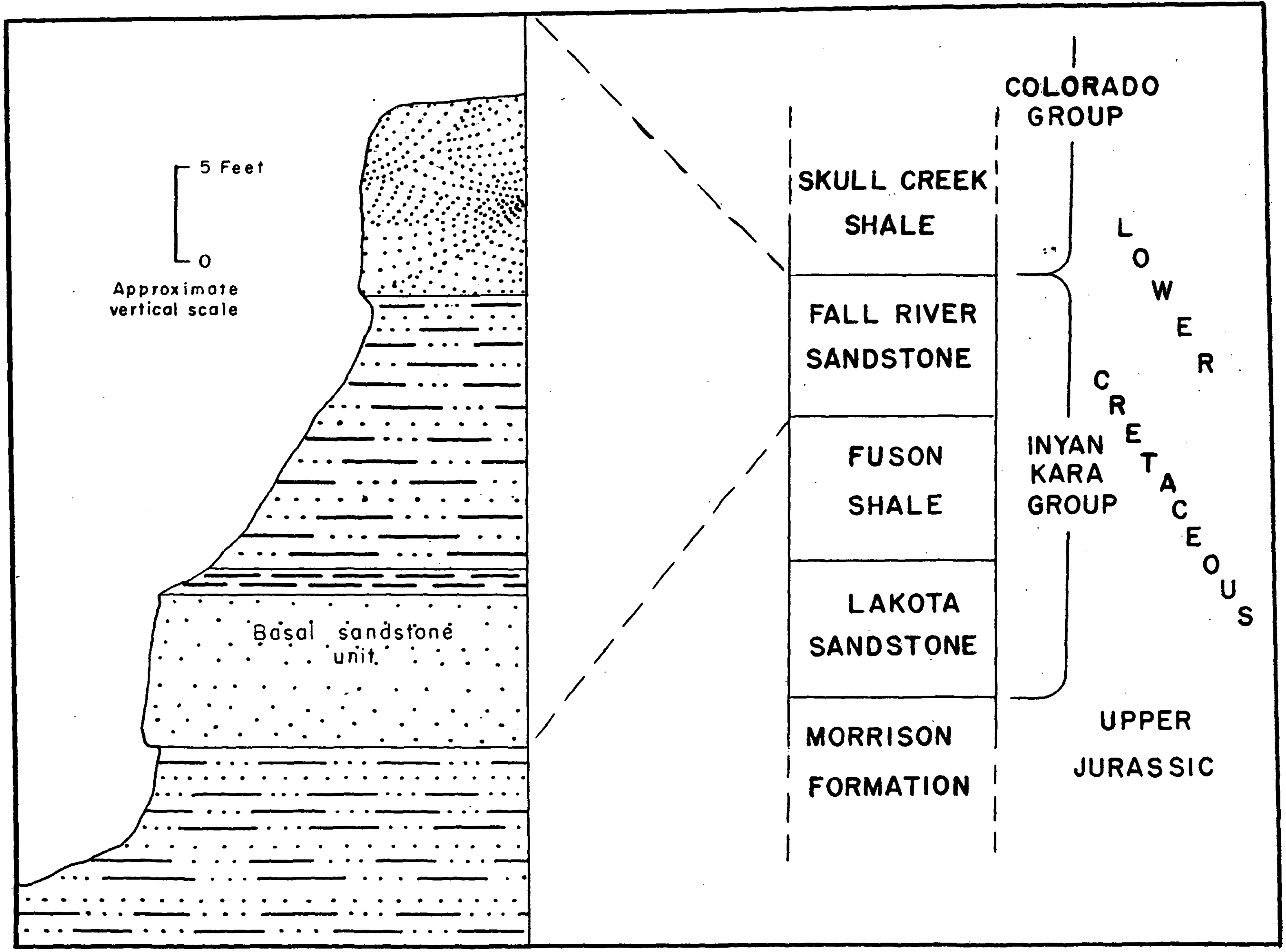


FIGURE 2.-GENERAL STRATIGRAPHIC SECTION, AND A TYPICAL CROSS SECTION IN AREA OF INVESTIGATION, BUTTE COUNTY, SOUTH DAKOTA

of the Fall River sandstone is well exposed and consists of 32 feet of gray to buff siltstone and shale that contains a few thin beds of limestone from one-half inch to five inches thick near the top. The contact with the overlying dark-gray fissile Skull Creek shale was not observed. The thickness of the Fall River sandstone ranges from about 35 to 55 feet, depending largely on the thickness of a bluff to reddish-brown cross-bedded sandstone as much as 30 feet thick that in places is present in the upper part of the formation.

Uranium deposits

All the uranium occurrences in the mapped area are in the basal sandstone unit of the Fall River sandstone except at the Bonato mine, where small quantities of carnotite were found in sandstone immediately above the shaly lignite which overlies the basal strata. However, the carnotite above the basal sandstone unit was localized along joints (oral communication from Clyde Boyle, mine operator) and may have been derived from the underlying sandstone by solution and redeposition.

Carbonized wood fragments are conspicuous at most of the carnotite occurrences. Carnotite is present as small pulverulent masses, as much as one-fourth inch in diameter, in carbonized wood fragments and as disseminations interstitial to, or coating, sand grains. The disseminated carnotite is commonly present in thin seams parallel to the bedding. The identification of the carnotite was confirmed by X-ray powder photographs.

With the exception of the Kling No. 2 mine (fig. 3), where about 450 tons of ore was removed, all the uranium occurrences are small, and only a few tons of ore-grade material were evident from most of the surface exposures.

ALTERATION OF SANDSTONE

Pink sandstone associated with uranium deposits was first recognized by Bales (Bell and Bales, 1955, p. 221) in the southern Black Hills. In general, the pink sandstone is there restricted to the immediate vicinity of the uranium deposits; but, in the northern Black Hills, in the area of this investigation reddish Fall River sandstone underlies several square miles or perhaps tens of square miles. Although it was realized from the previous work of Bell and Bales that the red sandstone might have some relationship to the uranium occurrences, the nature of this relationship was not appreciated until detailed mapping of the area had started. It then became apparent that most of the known uranium occurrences and radioactivity anomalies were in buff to gray sandstone immediately down dip from reddish altered sandstone. Both the buff and red sandstone are within the basal sandstone unit of the Fall River sandstone.

Nature of the red-buff contact

In most of the mapped area (fig. 3) the color of the basal sandstone unit of the Fall River formation changes from gray (yellowish gray, 5Y 7/2; to grayish yellow, 5Y 8/4) [1] to reddish (grayish orange pink, 5YR 7/2; pale red 10R 6/2; to pale reddish brown, 10R 5/4) within a horizontal distance of several feet or a few tens of feet. Commonly the contact between dominantly red and dominantly buff sandstone can be located within about 30 feet. Size analyses of both the red and buff sandstones at the Kling No. 2 mine showed no significant differences. A cross-section of a typical red-buff contact in the basal Fall River sandstone is shown in figure 4. The more intense red color of the altered sandstone is near the red-buff contact.

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[1] Rock Color Chart designations, National Research Council.

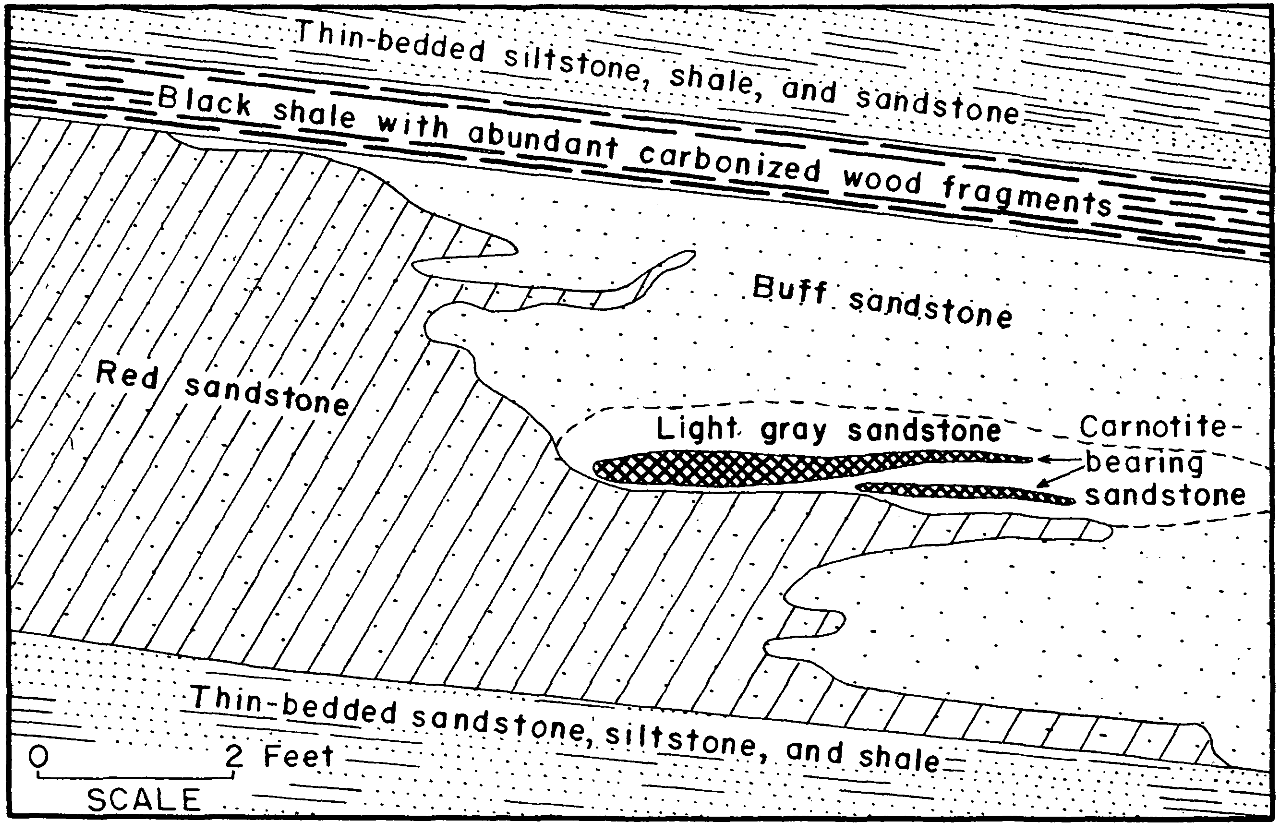


Figure 4.-Cross-section showing relationship of red-buff contact to carnotite-bearing sandstone.

Reddish color (pale red) was observed in the basal Fall River sandstone continuously for more than 1 mile to the southwest up dip from the red-buff contact. Reddish sandstone of the same sandstone unit was also observed a few miles both to the northwest and to the southeast of the mapped area.

In detail the reddish coloration of the sandstone at the red-buff contact cuts across the bedding and commonly extends farther down dip near the base of the sandstone. The red sandstone is believed to represent altered buff sandstone for the following reasons:

a. The basal Fall River sandstone is predominantly buff to gray where it is exposed on the northeast flank of the Black Hills (from Rapid City to Belle Fourche).

b. The red-buff contact cuts across the bedding and the red sandstone is a uniform color as contrasted with the color differences of adjacent beds in the buff sandstone.

c. At several localities in the mapped area, color bands of various shades of red are parallel to the red-buff contact, and the more intense red color is near the red-buff contact.

Mineralogy of the red and buff sandstones

Laboratory study of the Fall River sandstone suggests that the red color is probably the result of the alteration of ferrous iron minerals and yellowish brown hydrous iron oxides to red hematite.

The relative total iron content in each of several samples of red, buff, and gray sandstone was determined by X-ray fluorescence, and the red color is not a function of the total iron present. The total iron present in the samples ranges from 0.1 to 0.5 percent. (Spectrographic analyses by K. E. Valentine and N. M. Conklin, U. S. Geological Survey).

Samples of buff and gray sandstone were heated with a Meeker burner to about 600° C. for 20 minutes. The red colors produced by the heating were almost identical to the red colors in the natural red sandstones. This suggests that the red color in the basal unit of the Fall River sandstone resulted from the alteration of hydrous ferric oxides, and ferrous iron compounds, to hematite. Heavy-mineral determinations show abundant magnetite in the heavy minerals from the buff sandstone, whereas practically no magnetite was found in samples of red sandstone. With the exception of the iron oxides, no differences in mineralogy were noted between samples of nonuraniferous buff and red sandstone.

Relationship of uranium mineralization to the red-buff contact

All the uranium occurrences that have been found in the area of this investigation are immediately adjacent to the red-buff contact and with one exception are in buff sandstone. Three previously unknown occurrences of carnotite were found on the basis of the red-buff color change. Abnormal radioactivity (greater than 10 times background) was detected with a scintillation counter at practically all outcrops of the basal sandstone unit of the Fall River sandstone that showed the red-buff contact. About 32 radioactive localities were found at these outcrops, which are distributed along the sinuous contact between red and buff sandstone for about 5 miles. Abnormal radioactivity was also detected at a red-buff contact in Fall River sandstone about 1 mile northwest of the mapped area on the north side of Hay Creek valley. Southeast of the mapped area for at least 2 miles the red-buff contact was not observed; all outcrops noted of the basal unit of the Fall River formation were red.

Some of the radioactivity anomalies shown in figure 3 appear to be in the underlying Fuson shale; but this is caused mainly by radioactive float below outcrops of the basal sandstone.

Relationship of the red-buff contact to structure

As shown in figure 3, the red-buff contact in general parallels the strike of the regional dip but in detail cuts across the smaller structures. Mapping in the area indicates that small structures have exerted little control of the red coloration in the sandstone. An exception to this is in the SW¼ sec. 25, where unaltered buff sandstone occupies a structural trough that trends northeast.

Relationship of thickness to red-buff contact and uranium deposits

The thickness of the basal sandstone unit of the Fall River sandstone was measured at most outcrops where the full thickness was exposed. In the northwestern half of the mapped area, which includes all of the Kling deposits, no relationship of thickness to the uranium occurrences is apparent. The basal sandstone in this area is from 8 to 13 feet thick. In the southeastern half of the mapped area, many of the radioactivity anomalies and all the carnotite occurrences (except at the Bonato mine) are in sandstone generally less than 5 feet thick. Exposures are commonly poor where the basal sandstone unit is thin, therefore its thickness could only be approximated in many places.

In general, where the basal sandstone unit is thick (more than about five feet) the red-buff contact is found farther down dip than in areas where the sandstone is thin (less than about five feet). This relationship would be expected if ground waters moving down dip have caused the alteration.

Relationship of uranium deposits to structure

Minor structural features may have localized some of the uranium deposits. At the Kling No. 4 mine there is a local flattening of the regional dip; and for about 100 feet, measured perpendicular to the regional dip, there is a slight reversal of dip. The Kling No. 2 mine, about 200 feet down-dip from the Kling No. 4 mine, and the Kling No. 3 mine are also on small structural terraces. Several of the other uranium occurrences appear to be where the dip is less steep or are immediately down-dip from such areas.

The relationship between uranium deposits and structure is not marked, and the structural relationship noted above may be fortuitous. However, it may be significant that the largest uranium deposit (Kling No. 2) and the most widespread high radioactivity are down dip from the only observed reversal of dip found near the red-buff contact. Other uranium deposits of comparable size may not be present because other favorable structures are lacking.

ORIGIN OF THE URANIUM DEPOSITS

The red alteration described in this report may have significance relative to the origin of the uranium deposits. If we assume that the uranium was deposited from the same solutions that produced the red alteration--and the close association of the uranium with the red-buff contact strongly suggests this--then the uranium-bearing solutions moved generally down-dip. This direction would also be expected of normal ground-water flow in the area.

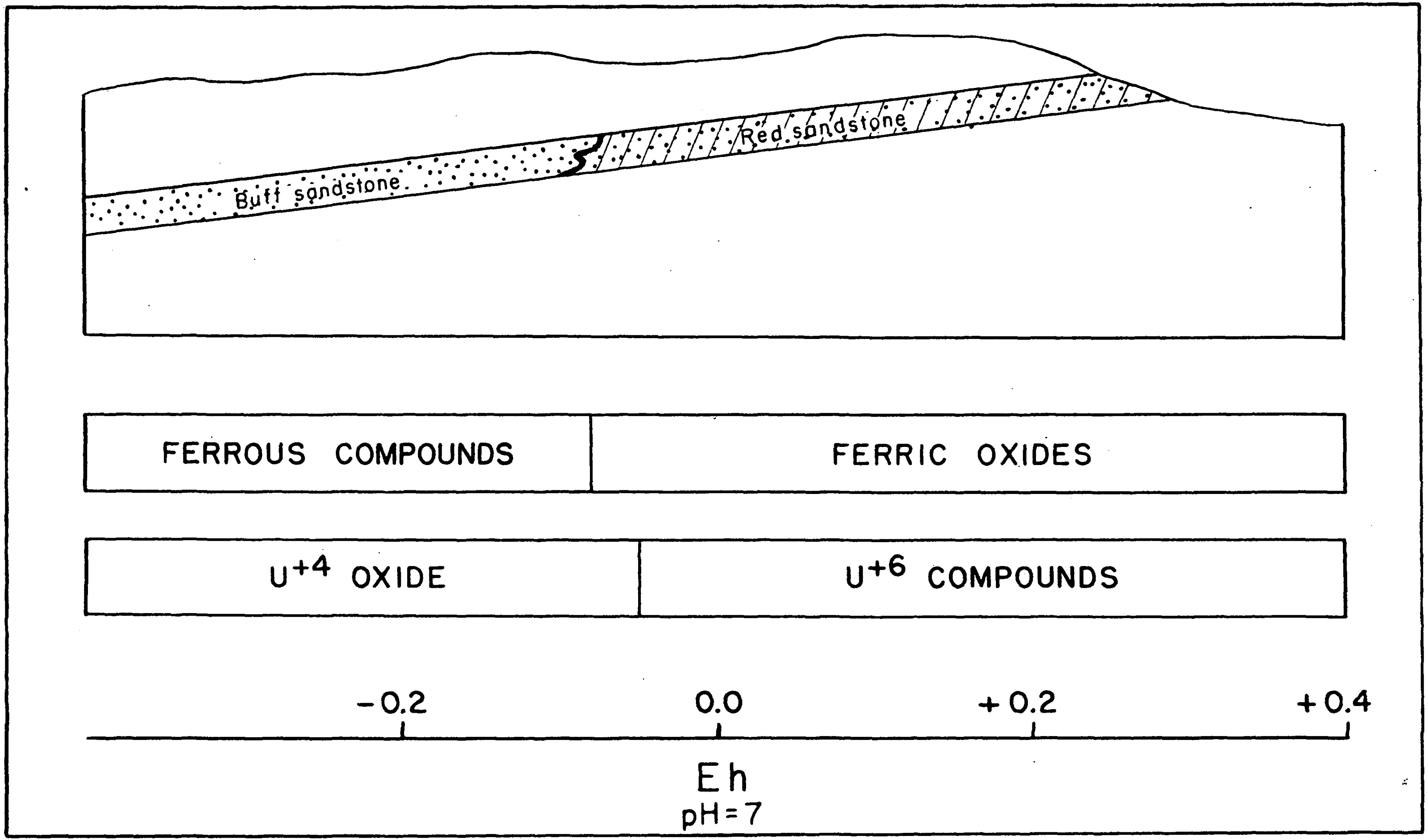
Because most of the uranium was deposited not in the red sandstone but immediately down-dip from the red-buff contact, alteration and uranium deposition probably occurred under different chemical conditions. Both field and laboratory evidence suggest that the uranium was deposited as pitchblende from downward-moving solutions that changed from a strongly to weakly oxidizing an even to mildly reducing character. Oxidation of the pitchblende during the present weathering cycle has resulted in the formation of carnotite and possibly other secondary uranium minerals.

The stability fields of uranium and iron minerals are consistent with this theory. Figure 5 shows the fields of stability of uranium and iron minerals as applied to the red-buff contact. These relationships suggest that pitchblende or perhaps coffinite was the original stable uranium mineral deposited because U^{+4} oxide (pitchblende) would be the stable uranium mineral in the buff sandstone that contains dominantly ferrous iron minerals below the zone of oxidation.

A laboratory experiment was conducted to determine the oxidation potential (Eh) and pH of waters in equilibrium with samples of both the buff and red sandstones. The samples of sandstone were sealed in glass containers with sufficient water to cover the samples. The Eh and pH of the solutions were measured after a period of six months with the following results:

	pH	Eh (volts)
Buff sandstone	6.80	+.084
Red sandstone	6.85	+.370

The above results, which show a much lower oxidation potential in the buff sandstone than in the red sandstone, are consistent with the theory of origin outlined above and suggest that natural waters in equilibrium with



Stability fields modified from Garrels, 1955

FIGURE 5. - FIELDS OF STABILITY OF URANIUM AND IRON AS APPLIED TO THE RED-BUFF CONTACT

the red and buff sandstones would have similar relative oxidation potentials. Although the Eh of the buff sandstone is not within the theoretical stability field of pitchblende, the samples of sandstone were taken from weathered outcrops and therefore the Eh values obtained from the buff sandstone may be considerably higher than those existing below the zone of oxidation.

The explanation for the lower oxidation potential in the buff sandstone is not completely known but is probably due in part to the larger amounts of ferrous iron in the buff sandstone than in the red sandstone. Carbonaceous material was not observed in the red and buff sandstone samples and therefore carbonaceous material is probably not responsible for the lower Eh in the buff sandstone.

Several experiments were conducted to determine the probable cause of environments sufficiently reducing to precipitate picthblende in sediments. Samples of water-saturated carbonaceous material from the Fall River sandstone that were sealed in glass containers showed relatively high Eh values after a period of from 1 to 18 months. However, samples of water-saturated pyrite from the same carbonaceous material produced very low Eh values. The results are tabulated below:

	pH	Eh (volts)
Carbonaceous material with most of pyrite removed	4.1	0.436
Pyrite from above carbonaceous material	6.6	-0.040

These results, although preliminary, suggest that pyrite may be a primary control for the localization of uranium deposits and that the association of carbonaceous material with uranium deposits may be due to the reducing environment caused by pyrite contained in the carbonaceous material. It is of interest to note in the above table that the pH and Eh values for the water-saturated pyrite are within the theoretical stability field of pitchblende as determined by Garrels (1955, p. 1017).

Although pyrite was not observed in specimens of carbonaceous material or sandstone from the Fall River sandstone in the area of investigation, pseudomorphs of goethite after pyrite were found in the basal Fall River sandstone, and abundant secondary gypsum, possibly derived from pyrite, is present in the weathered outcrops of carbonaceous beds. Pyrite is present in unoxidized and unmineralized drill-core samples of Fall River sandstone from several areas in the northern Black Hills.

Several different geologic conditions may have been effective in producing a sufficiently reducing condition for the precipitation of pitchblende. Ground water moving downward along a permeable bed would become more reducing through oxidation of various elements with which it comes in contact. This usual change from oxidizing to reducing conditions would in many cases occur over a large area and therefore would not be effective in localizing and concentrating uranium deposits. Localization of the reducing environment, to produce concentrations of pitchblende, would be necessary and could be caused by structural features such as terraces or small domes. On these structures, if the permeable bed were not saturated, there might be a local decreased rate of flow which would allow time for the solution to reach equilibrium with pyrite or other ferrous iron minerals and to become sufficiently reducing to precipitate uranium. A continual flow of water through the rock would essentially dilute the reducing capacity of any reducing material present; but if the flow were hindered or stopped, an area could be formed, within which conditions were sufficiently reducing for the reduction of hexavalent uranium. Local thinning of the sandstone or decrease in permeability could also inhibit the flow of ground water. Uranium ions moving into such an environment would then be deposited as pitchblende. Concentrations of carbonaceous material, especially those containing abundant pyrite, could cause localized reducing conditions.

The information concerning the origin of the uranium occurrences in the area of this investigation is summarized below:

1. The uranium-bearing solutions are believed to have moved generally down dip.

2. Alteration of buff sandstone to red sandstone is believed to have been nearly contemporaneous with the uranium deposition and this suggests that primary uranium was deposited during the change of the solutions from oxidizing to reducing.

3. Laboratory evidence suggests that ground water in equilibrium with the red sandstone has a much higher oxidation potential than ground water in equilibrium with the buff sandstone.

4. Concentrations of primary uranium have resulted from localizations of reducing conditions. (Carnotite has formed by oxidation of the primary uranium during the present weathering cycle.)

5. Pyrite-bearing sandstone and pyritiferous carbonaceous material are effective in producing an environment sufficiently reducing for the precipitation of pitchblende.

6. Structures such as terraces or domes superimposed on the regional dip, local thinning or decrease in permeability, or concentrations of pyritiferous material may have been effective in localizing the reducing environment.

The evidence cited above justifies the conclusion that the uranium deposits have formed by the action of ground waters. However, the origin of the uranium in the ground water presents a separate problem and one which is closely related to the origin of the red altered sandstone.

ORIGIN OF THE RED ALTERATION

The red altered sandstone is probably the result of pre-Oligocene weathering. An altered zone below the base of the Oligocene has been recognized by several workers. Tourtelot (1956, p. 80) states:

> "Throughout large areas of the Great Plains in Nebraska and South Dakota, the predominantly shaly marine strata of Late Cretaceous age are overlain by tuffaceous sandstone and claystone of the Chadron formation at the base of the White River group of Oligocene age. A conspicuous feature of the unconformable contact between these two sequences is a brightly colored zone of altered Cretaceous shales beneath the Chadron.
>
> The zone is as much as 50 feet thick and is present everywhere except where channels were cut through it prior to the deposition of the Chadron formation. The altered shale ranges in color from nearly white through shades of yellow to orange, brown, and purple. Red streaks are common, and oxides of iron coat joints and bedding planes."

After detailed work on the weathered zone, Dunham (1955, p. 8) found that the upper part of the weathered zone exhibits the chemical, mineralogical, and textural characteristics of a maturely developed soil.

Although rocks of White River age are not present in the immediate vicinity of the area of investigation, small outliers of pebble conglomerate, believed to be of White River age, rest unconformably on the Fall River formation in several localities in the northern Black Hills; in the area of investigation the pre-White River erosion surface was probably only about two hundred feet above the present bedrock surface. The red-buff contact may be the extension, down dip in the more permeable beds of the lowermost zone of a soil profile which was developed on pre-Oligocene strata.

Red alteration is not restricted to the Fall River sandstone in the northern Black Hills. For example, the Hulett sandstone member of the Upper Jurassic Sundance formation is also altered red for a considerable distance down-dip from the pre-White River erosion surface that was locally developed on the Hulett sandstone on the north and west flanks of the Bear Lodge Mountains. The Bear Lodge Mountains are about 20 miles southwest of the area of investigation.

SOURCE OF THE URANIUM

The source of the uranium and associated elements is unknown. However, the author believes that these elements have been derived from dispersed sources in the sediments which have been eroded. Some of the uranium may have come from the altered part of the Fall River sandstone. There is no evidence to indicate that hydrothermal solutions have played a part in the origin of the uranium deposits. On the contrary, the available information concerning the behavior of uranium during weathering processes suggests that weathering of a slightly uraniferous rock coupled with a suitable reducing environment below can adequately account for the deposits.

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