The origin and properties of uranium–niobium–tantalum mineralised hydrocarbons at Narestø, Arendal, southern Norway

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The mineralogy, chemical and physical properties of uraniferous hydrocarbons (0.86 wt.% uranium) from a pegmatite at Narestø, near Arendal have been investigated. The hydrocarbon contains irregular inclusions of uraninite and grains of xenotime and monazite, relics of minerals formerly in the pegmatite mass. The uraninite has been corroded by reaction with the hydrocarbon. Aeschenyte and quartz fill fractures and form overgrowths around other minerals within the hydrocarbon. The properties of the hydrocarbon (specific gravity = 1.51–1.55, R_0 = 1.63, hardness = 5–6, H/C = 0.52–0.63 and $\delta^{13}C$ = −26.6‰ to −26.9‰ PDB) compare with those of thermally altered hydrocarbons from Kongsberg and Åmot. These differ only in having lighter stable carbon-isotopic compositions and lower H/C ratios. In comparison, thermally immature bitumens from Sweden have high H/C ratios (1.04–1.46), low specific gravities (1.0–1.2) and reflectances (R_0 = 0.01–0.17). The isotopic compositions of the Swedish bitumens compare with the non-uraniferous hydrocarbons from Norway. A biogenic source for the Narestø hydrocarbon is suggested on the basis of carbon-isotopic evidence.

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The occurrence of radioactive hydrocarbon in the feldspar quarry at Narestø near Arendal in southern Norway (Fig. 1) has been known since the last century (Helland 1875; reported in Dons 1956). The hydrocarbons are found within a granite pegmatite composed of alkali and plagioclase feldspar, quartz, biotite and magnetite. Some slightly radioactive minerals such as monazite, xenotime and orthite are also reported (Andersen 1931; Heinrich 1958). The pegmatite is one of a series developed within the Bamble formation of southern Norway and is assumed to be of Precambrian age (Lindahl 1983). Non-uraniferous hydrocarbons (known locally as coal blend) also occur at several localities in southern Norway (Fig. 1), including the Kongsberg silver deposit, and at Åmot, Modum where it is associated with a breccia zone, crosscutting Precambrian gneiss and schist intruded by Permian dykes. All three localities are described by Dons (1956). The Kongsberg and Åmot samples are associated with extensive calcite mineralisation and contain traces of vanadium. They may be of similar origin and age, being formed at elevated temperatures associated with late Palaeozoic rifting. Fluid inclusion studies indicate temperatures of formation between 200° and 400°C for the Kongsberg mineral veins (Ihlen & Vokes 1978). The coal blend-bearing stages of mineralisation predate the intrusion of dykes around the Oslo graben (Ihlen 1980). As a result the Åmot material may also have been heated by the intrusion of two Permian dykes exposed at the locality. In contrast, the Narestø hydrocarbons do not appear to have experienced the same elevated temperatures, as no igneous intrusions are known around the immediate vicinity of the occurrence, although dolerite dykes are known to have intruded the Arendal area. However, the physical and chemical properties of the hydrocarbons would have been affected by radiation emitted from uranium within the hydrocarbon. The effects of radiation over geological time have been likened to the effects of extreme thermal alteration (Landais & Connan 1986). Thus, the availability of samples of hydrocarbons from Narestø, Åmot and Kongsberg, supplied by the Mineralogical-Geological Museum of Oslo, realised the opportunity to compare the chemical and physical properties of hydrocarbons affected by
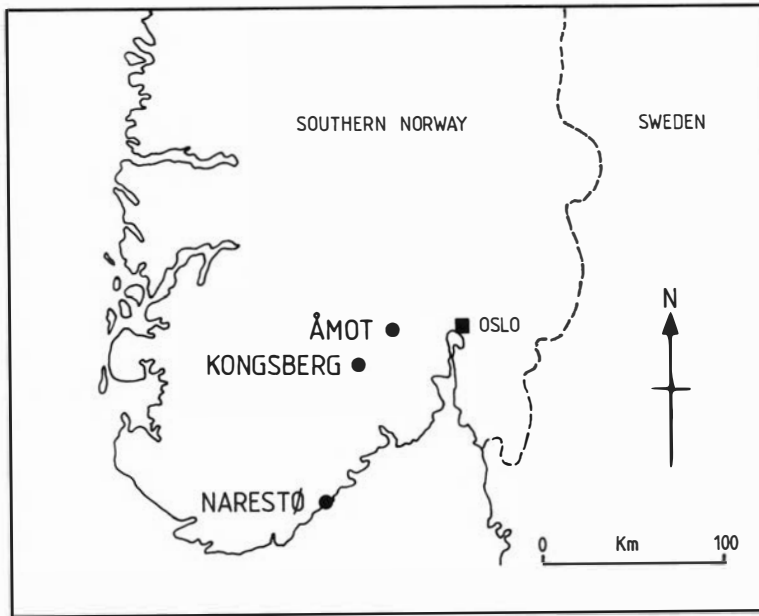
Fig. 1. Location map of the localities of Norwegian hydrocarbon samples mentioned in the text.

extreme thermal alteration (Kongsberg and Åmot) with those altered by radiation over geological time (Narestø). The results of these analyses were compared with those of a suite of non-uraniferous, low thermal maturity bitumens from four localities in Sweden.

In addition, back-scattered electron microscopy and microprobe analyses have been used to study the Narestø hydrocarbon in order to elucidate the geological history and provide evidence of the mode of formation of the material. This is an important question in the debate over the existence or otherwise of abiogenic hydrocarbons.
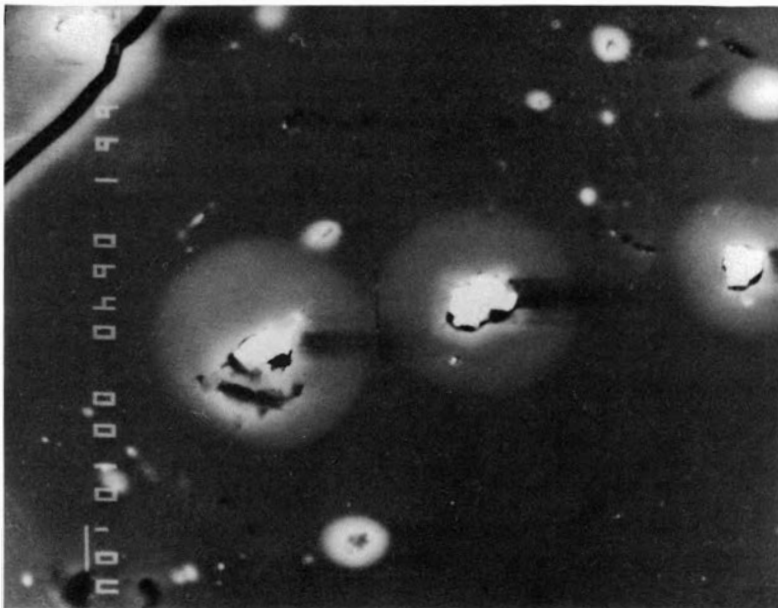

Fig. 2. Backscatter electron micrograph of irregular uraninite grains floating in hydrocarbon. The bright rings around the grains are due to radiation damage of the hydrocarbon (scale bar 10 μm). Dark streaks are artefacts of scanning technique.

Table 1. Physical and chemical properties of Norwegian hydrocarbons from Narestø, Åmot and Kongsberg, and four thermally immature, non-uraniferous hydrocarbons from Sweden

Sample	U (wt.%)	S.G.	Hardness (Moh's)	Lustre	R_0 (%) (in oil)	H/C Ratio	$\delta^{13}C$ (‰ PDB)
NORWAY							
Narestø							
1909	0.86	1.51	5–5.5	M	1.63	0.52	−26.9
1939	—	1.55	5–6	sM	—	0.63	−26.6
Kongsberg	—	1.63	3–4	sM-W	4.69	0.35	−30.2
Åmot	0	1.55	>6	M	3.98	0.09	−30.3
SWEDEN							
Granmuren	—	1.00	1–2	R	0.01	1.46	−29.4
Nyäng	—	1.31	—	1	—	1.29	−29.5
Norberg	—	—	—	R	—	1.34	−29.9
Pajsberg	—	1.20	—	R	0.17	1.04	−30.0

(M = metallic, sM = submetallic, W = waxy, R = resinous, 1 = liquid)

Analytical techniques

Back-scattered electron microscopy and micro-probe analyses were carried out on a Joel 733 Superprobe, analysis being performed using an accelerating voltage of 15 KeV and 5 μm probe diameter. Results were processed through Zaf 4 computer software.

Bitumen reflectances (R_0) were measured on a Zeiss UMSP 50 light system connected to an IBM PC computer. Reflectance measurements were made in oil immersion (R.I. oil = 1.515) using a ×40 objective lens, 0.16 mm measuring disc, 0.32 mm field width and monochromatic light (wavelength 546 nm). Precision and accuracy of reflectance measurements are better than 0.04% based on the determination and repeated analyses of an internal standard (R_0 = 1.73%).

Samples for chemical analyses were crushed in an agate mortar and pestal. The stable carbon-isotopic compositions of the hydrocarbons were measured using the carbon oxidation method described by Sofer (1980). The purified CO_2 was analysed on a VG Isogas Sira 10 mass spectrometer and the results computed relative to the Pee Dee Belemnite standard. Precision and accuracy are better than 0.15‰. Uranium determinations were made by delayed neutron activation analysis; elemental analyses were also performed. In addition, specific gravity (S.G.), hardness and lustre were determined for each sample.

Results

Neutron activation analysis reveals that the Narestø hydrocarbon contains 0.86 wt.% uranium (Table 1). The back-scattered electron microscopy/microprobe studies of the hydrocarbon show that the uranium is concentrated in a distinct mineral phase 'floating' in hydrocarbon. The mineral inclusions range in size from 5 to 50 μm in diameter and have irregular boundaries, which reflected light microscopy reveals to be highly altered. The grains commonly occur as closely packed aggregates, with hydrocarbons filling the intervening spaces. Probe analyses of these minerals (Table 2) show them to contain up to 86.8% UO_2, with traces of PbO, ThO_2 and SiO_2. The inclusions have been identified as uraninite by X-ray diffraction.

Other minerals found in the hydrocarbon

Table 2. Electron-microprobe analyses of uraninite grains in the Narestø hydrocarbon. (Oxygen is calculated by stoichiometry and the results normalised to 100%).

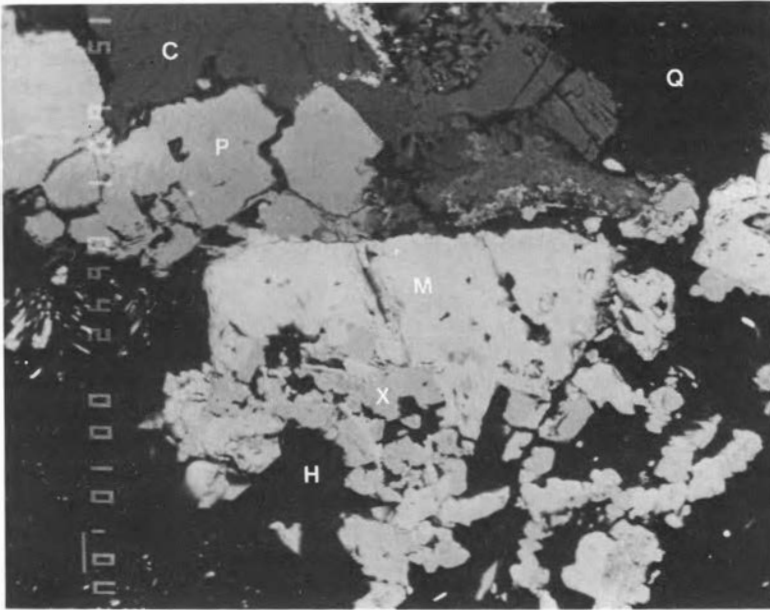| UO_2	ThO_2	PbO	SiO_2	Total (wt.%)
83.39	7.15	9.26	0.24	100.04
84.59	7.88	6.33	0.32	99.12
77.71	6.62	8.52	7.19	100.04
86.00	4.86	3.20	5.89	99.95
80.47	7.45	10.41	1.69	100.02
86.82	5.83	3.64	3.72	100.01

Fig. 3. Back-scatter electron micrograph of complex intergrowths of xenotime (X), monazite (M), pyrite (P), cacite (C) and quartz (Q) in hydrocarbon matrix (H); (scale bar 10 μm).

include complex but uncorroded intergrowths of the phosphates xenotime and monazite (Fig. 3), which are slightly radioactive, containing up to 1.3% UO_2. Qualitative probe analysis indicates the presence of thorium, yttrium, and some rare-earth elements in the phosphates.

A third suite of minerals fills fissures 5–20 μm wide and of variable length, and forms over-growths around other mineral grains within the hydrocarbon (Fig. 4). These minerals include a phase rich in niobium, tantalum, barium and uranium, with traces of yttrium, thorium, titanium and iron, which yields a general formula of $(Ba, U, Y, Th, Ti, Fe)_{1.2-1.6}(Nb, Ta)_2(O, OH)_6$.

Fig. 4. Back-scatter electron micrograph of Nb–Ta minerals filling veins and fractures in the hydrocarbon (scale bar 100 μm).
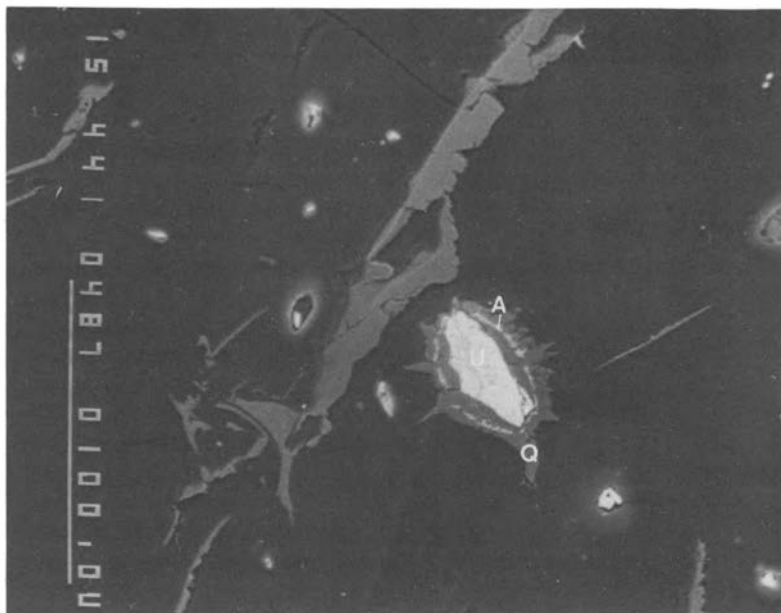
Fig. 5. Back-scatter electron image of quartz veins cutting hydrocarbon and forming a zoned overgrowth (Q), with aeschynite (A) on uraninite (U); (scale bar 100 μm).

It is tentatively identified as a barium-rich form of aeschynite (U, Th, Ce, Ca, Fe) (Nb, Ta)$_2$(O, OH)$_6$ (Morton 1978). Quartz is also present in this suite, but post-dates the aeschynite (Fig. 5). Pyrite and galena also occur as inclusions in the hydrocarbon mass (Fig. 3).

Back-scattered electron imagery also reveals the presence of halos in the hydrocarbon surrounding the uraninite, which appear brighter than the hydrocarbon not in the vicinity of the uraninite grains (Fig. 2).

The results of the chemical and physical property determinations on the Norwegian and Swedish hydrocarbons are shown in Table 1. The non-uraniferous hydrocarbons from Sweden have properties and chemical compositions typical of bitumens of low thermal maturity, which contrast with those from Narestø, Åmot and Kongsberg. The Swedish samples have high H/C ratios, low specific gravities, reflectances less than 0.17%, and are soft or liquid in character. The $\delta^{13}C$ ratios of the samples fall in the narrow range of −29.4‰ to −30.0‰ PDB.

The Narestø hydrocarbon is a black to dark grey, brittle substance with a conchoidal fracture, yielding a dark grey streak. The lustre is metallic to submetallic, the hardness ranges from 5 to 6 (Moh's scale) and the specific gravity from 1.51 to 1.56. These properties compare closely with those of the hydrocarbons found at Åmot and

Kongsberg, which have similar appearance, specific gravity and hardness. However, the atomic H/C and $\delta^{13}C$ ratios of the Åmot and Kongsberg samples differ substantially from those of the Narestø samples. The carbon-isotopic ratios of the non-uraniferous samples are similar to those of the Swedish samples (−30.2‰ to −30.3‰ PDB) and are isotopically lighter by approximately 3‰ compared with the uraniferous sample. The H/C ratios of the thermally altered hydrocarbons are also markedly less than the thermally unaltered Swedish samples.

There is substantial variation in bitumen reflectance both within the uraniferous hydrocarbon, and between the Narestø sample and the thermally altered hydrocarbons. The mean reflectance values (in oil) for the bitumens are shown in Table 1 and indicate greater bitumen reflectance in the Åmot and Kongsberg material ($R_0 =$ 3.98% and 4.69% respectively) compared with the Narestø samples ($R_0 = 1.7\%$). The variation in reflectance within the Narestø sample arises from the increase in reflectance of the bitumen in close proximity to the uraninite inclusions, the reflectance gradually increasing towards the grains (Eakin 1989). A plot of uraninite grain diameter against the bitumen reflectance in the immediate vicinity of the inclusion shows a positive linear correlation between the two parameters (Fig. 6).
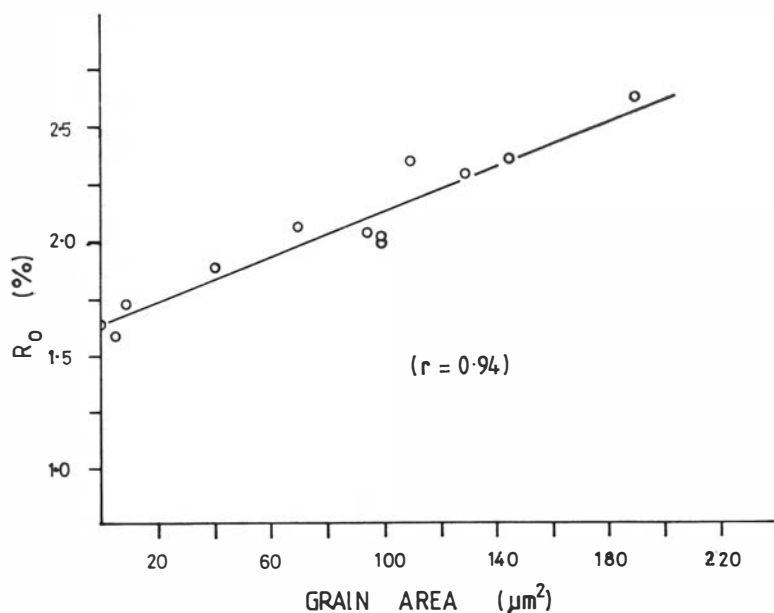
Fig. 6. Plot of uraninite grain area exposed in polished section against the mean bitumen reflectance of the bitumen in the immediate vicinity of the uraninite grain.

Discussion

The early descriptions of thucholites (uranium- and thorium-rich hydrocarbons) and uraniferous hydrocarbons in pegmatites were from examples in the pegmatite dykes of Ontario, Canada (Ellsworth 1928; Spence 1930). Early workers considered the association of hydrocarbons and pegmatites to be evidence for the existence of hydrocarbons of an abiogenic origin (e.g. Ellsworth 1928).

The granite pegmatite at Narestø is developed within Precambrian gneiss locally crosscut by Permian dykes. The dykes contain vesicles filled with oil, which led to the suggestion that the hydrocarbons had an inorganic origin, formed as a product of the reactions between carbon dioxide and water at elevated temperatures (Evans et al. 1964).

The Lower Palaeozoic strata of Scandinavia, which could have covered the Arendal area at one time, include the kerogenous upper Cambrian Alum shales and many other dark shale members (Bergström & Gee 1985). These are viable oil source rocks, and are thought to have contributed to many minor oil and bitumen occurrences in the Precambrian basement rocks of Scandinavia (Welin 1966; Andersson et al. 1985).

Thus, an alternative theory for the genesis of the hydrocarbons in the intrusions involves the heating of these shales during Permian igneous activity, and the migration of hydrocarbons thus produced into the intrusions. Based on the carbon-isotopic composition of the uraniferous organic matter (see below), a biogenic origin for the Narestø hydrocarbons is preferred here.

The results of the elecron-microscopy investigations on the hydrocarbon suggest several stages of mineral development within the hydrocarbons. It is assumed that the uraninite in the hydrocarbons represents relics of crystals originally present in the granite pegmatite. The high percentage of thorium in the uraninite (Table 2) is consistent with the formation of the uraninite at the elevated temperatures associated with pegmatite development (Durrance 1986). The irregular shape and the isolation of the uraninite grains and grain aggregates are suggestive of corrosion by the hydrocarbon. Such alteration has been suggested by many authors to account in part for the common association of uranium and organic matter (e.g. Davidson & Bowie 1951; Hoekstra & Fuchs 1960; Miller & Taylor 1966; Schidlowski 1975). Substantial corrosion of clastic grains associated with uraniferous hydrocarbons has also been documented (Parnell & Eakin 1987).

Hydrothermal systems, formed in response to the intrusion of the Permian dykes, may have been responsible for the migration of hydrocarbons from the dykes to the site of deposition in the Narestø pegmatite. Here, radiation-induced polymerisation reactions may have caused the precipitation of hydrocarbons around, with simultaneous corrosion of, the uraninite in the pegmatite.

The presence of niobium–tantalum mineralisation supports the theory that hydrothermal fluids were responsible for the movement of both hydrocarbons and other elements to the site of mineralisation in the pegmatite. Niobium and tantalum are mobile during hydrothermal activity (Beus & Sitnin 1961) and may have had their source in Nb/Ta minerals found in pegmatites within the Bamble formation of southern Norway (Oftedal 1972). In addition to the precipitation of aeschenyte, minor amounts of sulphides were also formed, and quartz veinlets and overgrowths were developed during the later stages of the hydrothermal activity, after the formation of the uraniferous hydrocarbon.

The traces of uranium within the aeschenyte may be due to uranium released into the hydrocarbon during the corrosion of the uraninite by the organic matter, and then incorporated into the Nb/Ta mineral during its precipitation. Uranium-enriched minerals commonly fill fractures within uraniferous hydrocarbons assumed to be formed by the corrosion of pre-existing uraninite and pitchblende by hydrocarbons. For example, trögerite, a uranium/arsenic mineral, has been found filling fractures in hydrocarbons corroding vein-hosted pitchblende (Parnell 1988), and brannerite occurs within hydrocarbons corroding uraninite grains (Schidlowski 1975) in the 'Carbon Leader' uraniferous hydrocarbons of the Witwatersrand region of South Africa.

The volume increase which accompanies the corrosion of uraninite by hydrocarbon (Hoekstra & Fuchs 1960) may be responsible for the incorporation into the hydrocarbon of unaltered xenotime and monazite derived from the pegmatite. This process could occur as hydrocarbons are forced into small cracks and fissures in the pegmatite, causing small amounts of mineral to be separated from the main mass and incorporated into the hydrocarbon.

The physical and chemical properties of the hydrocarbons from Sweden are typical of those of low thermal maturity and uranium content (Table 1). They are characterised by high H/C ratios (>1), low reflectance and specific gravity, and have the appearance and properties of bitumen or hardened oil. The samples from Kongsberg and Åmot are typical of hydrocarbons of high thermal maturity, being marked by low H/C ratios and higher specific gravity, reflectances and hardness. The similarity between the carbon-isotopic compositions of the thermally mature hydrocarbons and those from Sweden is of note, all the values falling in the narrow range between −29.4‰ and −30.3‰ PDB. This isotopic composition is typical of organic compounds of biological origin (Deines 1980). The lack of variation between the samples is suggestive of a common hydrocarbon source, for example the lower Palaeozoic shales mentioned above. The lack of variation between the thermally mature and immature samples is consistent with the findings of Chung & Sackett (1979), who noted only a 0.5‰ change in the $\delta^{13}C$ ratios of organic matter heated to 500°C during experimental pyrolysis.

The markedly heavier stable carbon-isotopic composition of the uraniferous hydrocarbons is consistent with the theory of radiation-induced fractionation of stable carbon isotopes in uraniferous organic matter (Leventhal & Threlkeld 1978). Eakin (1989) noted a +3‰ fractionation in uraniferous hydrocarbons from Great Britain, of similar uranium content to that of the Narestø hydrocarbon. If fractionation has occurred in the Narestø hydrocarbon, the original hydrocarbon could have had a stable isotopic composition similar to that of the other Scandinavian samples (approximately −30‰ PDB), supporting the suggestion that the Narestø hydrocarbons had the same (organic) source as the hydrocarbons from Sweden, Åmot and Kongsberg. The alternative (abiogenic) source, hydrocarbons formed by the reaction of CO_2 with water in the dolerite dykes of the Arendal area (Evans et al. 1964), would require the incorporation of carbon from either an atmospheric or mantle source. These carbon reservoirs have mean isotopic compositions of −5‰ and −7‰ respectively (Schidlowski 1988). Thus, a fractionation of approximately −20‰ would have to be proven during the incorporation of CO_2 into the intrusions, the formation of the hydrocarbons and their transport to, and incorporation into, the uraniferous hydrocarbon of the pegmatite, before an abiogenic source within the dykes could be seriously considered for the Narestø hydrocarbon. It is difficult to envisage

how this could be achieved. However, firm conclusions on the origin of the hydrocarbons in the dolerite dykes must also wait until further investigations, including isotopic analysis, have been performed.

Other properties of the uraniferous hydrocarbons reflect the effects of radiation alteration on the hydrocarbon. The properties include higher specific gravity, lustre, hardness, and H/C ratios markedly lower than those of the Swedish bitumens (Table 1). These properties are typical of those of uraniferous hydrocarbons, compared to the characteristics of their non-uraniferous equivalents (Haji-Vassiliou & Kerr 1973). The properties are similar to those of the thermally altered hydrocarbons. The similarity of the specific gravities is surprising, given the uranium content of the Narestø hydrocarbon, which should increase the specific gravity of that sample. However, the markedly lower H/C ratios of the thermally mature hydrocarbons indicate that their specific gravity has increased during maturation as a result of substantial alteration by thermal cracking, volatile loss and graphitisation, as the hydrocarbons approach a graphitic composition, as indicated by the optical characteristics of the Kongsberg hydrocarbon (Gize 1985). Radiation-induced maturation of hydrocarbons involves volatile loss (Colombo et al. 1964) with a consequent reduction in H/C ratios (Charlesby 1954), but apparently did not cause the same extreme reduction in H/C ratio in the Narestø hydrocarbon as in the thermally altered Kongsberg and Åmot samples. This may be because the degree of radiation-induced alteration is not constant throughout the sample, but is maximised close to the uraninite grains, as indicated by the increase in bitumen reflectance close to the grains (Eakin 1989).

Increased reflectance of organic matter around uraniferous grains is well known (Breger 1974; Schidlowski 1975; Gentry et al. 1976; Leventhal et al. 1987). Figure 6 indicates that the increase in bitumen reflectance close to the grains is related to the size of the grain, supporting the theory that such high reflectance halos form as a result of the action of alpha radiation, emitted from the uraninite grain, on the organic matter (Leventhal et al. 1987; Eakin 1989). Radiation-induced reactions result in more condensed, aromatic organic matter formed by cross-linking, aromatisation and devolatisation reactions (Charlesby 1954; Colombo et al. 1964). Such reactions combine to increase the reflectance of the organic matter, and will be maximised close to the uraninite grains. These processes are also likely to be responsible for the formation of the halos visible in backscattered electron imagery of the hydrocarbon. This method of microscopy gives images of greater intensity for substances of greater atomic weight. Thus, the presence of the halos indicates that the hydrocarbon surrounding the uraninite grains has a different composition compared with that of the rest of the hydrocarbon mass. Parnell (1988) noted an increase in the oxygen content of hydrocarbons in similar halos around uraninite grains in hydrocarbons from the Isle of Man. Alternatively, the production of more graphitic hydrocarbon by the action of radiation may be sufficient to alter the intensity of the back-scattered image in the vicinity of the uraninite grains.

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