Magnesite deposits at Modum – how did they form?

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Introduction

The magnesite-serpentine deposits of the Modum area (Buskerud, South Norway) are well known to Norwegian mineral collectors. About 20 smaller and larger occurrences have been registered (Jøsang 1966), see Fig. 1. Mining for magnesite, to be used for basic furnace linings after calcination to magnesium oxide, was conducted in the period 1905–1960 at 11 localities, the largest and most famous mines being those at Dypingdal, Tingelstadtjern and Overntjern. The complete mineral inventories of these occurrences can be found at *mindat.org* and there is no need for repeating them here. The magnesite deposits at Modum are mineralogically unique, and even remotely similar occurrences seem not to have been observed elsewhere in the world.

Dypingdal is the type locality of two carbonate minerals:

Hydrotalcite, $Mg_6Al_2(CO_3)(OH)_{16} \cdot 4H_2O$ (Hochstetter 1842, Raade 2013)

Dypingite, $Mg_5(CO_3)_4(OH)_2 \cdot 5H_2O$ (Raade 1970, 2012)

A unique assemblage of new magnesium phosphates is described from Tingelstadtjern (Raade 1999):

Althausite, $Mg_4(PO_4)_2(OH,O)(F,\square)$ (Raade & Tysseland 1975, Rømming & Raade 1980)

Holtedahlite, $Mg_{12}(PO_3OH,CO_3)(PO_4)_5(OH,O)_6$ (Raade & Mladeck 1979, Rømming & Raade 1989)

Heneuite, $CaMg_5(CO_3)(PO_4)_3(OH)$ (Raade *et al.* 1986, Rømming & Raade 1986)

Phosphoellenbergerite, $Mg_{12}(Mg,Fe,\square)_2(PO_4,PO_3OH,AsO_4)_6(PO_3OH,CO_3)_2(OH)_6$ (Raade *et al.* 1998)

Raadeite, $Mg_7(PO_4)_2(OH)_8$ (Chopin *et al.* 2001; observed as an unknown phase by Raade *et al.* 1986)

A metamict, thorium-rich magnesium phosphate was described from Overntjern (Raade 2011).

Apart from having described five new minerals from these deposits, I have also considered their petrogenesis (Raade 1986). The paper is included in a book entitled *Metallogeny of Basic and Ultrabasic Rocks (Regional Presentations)*, published by Theophrastus Publications. This publishing company was run by Professor S. S. Augustithis of the National Technical University of Athens. It published a number of books on various geological topics in the 1970s to 1990s. Unfortunately, some of the books suffered from a limited distribution, and therefore my paper has not been seen by many. I am pleased to have this opportunity to give an extended summary of the results.

Previous investigations

Raade (1986) reviewed the earlier mineralogical literature on the magnesite deposits and showed that numerous papers were published in the 19th century but relatively little thereafter. Jakob Schetelig (1875–1935) collected a large number of mineral samples from the magnesite localities but did only

publish a short paper on two phyllosilicates (Schetelig 1920). Field work by Henrich Neumann in 1940 was recorded in unpublished reports. An extensive sampling of minerals was conducted by Ottar Jøsang in the years 1953 to 1956, from magnesite mines that were still in operation, along with a detailed mapping of the area. The geological and petrographical results were published (Jøsang 1966), but the mineralogical investigations were never completed. The present author has performed systematic mineral collecting at all deposits in 1976 and later and has published a preliminary account of geochemical work on these samples in the 1986 paper. It contains electron probe microanalyses (EPMA) of almost all of the minerals and trace-element compositions of some of them by instrumental neutron activation analysis (INAA). In addition, oxygen and carbon isotope compositions of magnesite were determined. The most important results of this geochemical work, which has bearing on the origin of the magnesite-serpentine rocks, are summarized in the following.

Geology of the area

The main rocks of the Modum Complex are amphibolitized gabbros (metagabbros) and amphibolites in large volumes, banded gneisses, micaschists, sillimanite granites, granitic pegmatites, quartzites, scapolite rocks and different sorts of albitites (Jøsang 1966, Munz & Morvik 1991). The episode of extensive albitization was explained as a result of retrograde fluid infiltration by Munz et al. (1994). Whiteschists and orthoamphibole-cordierite rocks are found in contact zones between metagabbros and supracrustals (Munz 1990). The general strike of the rocks is north–south, and the distribution of magnesite-serpentine bodies is seen roughly to follow this pattern (Fig. 1).

The serpentine bodies form steeply dipping, elongated lenses up to 100 m long and 20 m wide. They consist mainly of alternating layers of ophimagnesite (a mixture of magnesite and serpentine) and serpentine. Pure, saccharoidal magnesite is rare, but smaller volumes of coarsely crystalline magnesite are found in most of the occurrences. Tectonic influence is evidenced by numerous slickensides, bulging cleavage planes in magnesite and althausite and development of glide twinning on $\{10\bar{1}1\}$ in hematite. Brecciation of the surrounding rocks seems to be a contemporaneous event. The lensoid bodies of serpentine and magnesite usually have concordant contacts with neighbouring metamorphic rocks.

Theories on the origin of the Modum magnesite deposits

Owing to their economic importance and widespread occurrence worldwide, the origin of magnesite deposits has been extensively studied. The majority seems to have originated as sediments, and some have had a magmatic precursor (Pohl & Siegl 1986). As for the Modum magnesite deposits, thoughts on their primary nature have been briefly expressed by several authors. Schetelig, in a lecture given in 1925, conveyed the view that they were undoubtedly of magmatic origin, designating them as "dunite pegmatites" (Schetelig 1926). This conclusion was mainly based on mineral assemblages. Bugge (1936, 1937) has used the term "circulating fluids rich in MgO and CO_2" and seems to favour a hydrothermal origin. Based on the occurrence of forsterite pseudomorphs, Faust & Fahey (1962) have classified the Dypingdal deposit as an altered ultramafic body. The careful mapping done by Jøsang (1966) led him to assume that the precursor rocks could have been sedimentary dolomites. He observed that anthophyllite-bearing quartzite remarkably often forms the wall-rock of the magnesite-serpentine bodies, alternately along the hanging wall or the footwall. This feature was interpreted to reflect the presence of a folded metasedimentary sequence in which these rocks occupy a specific
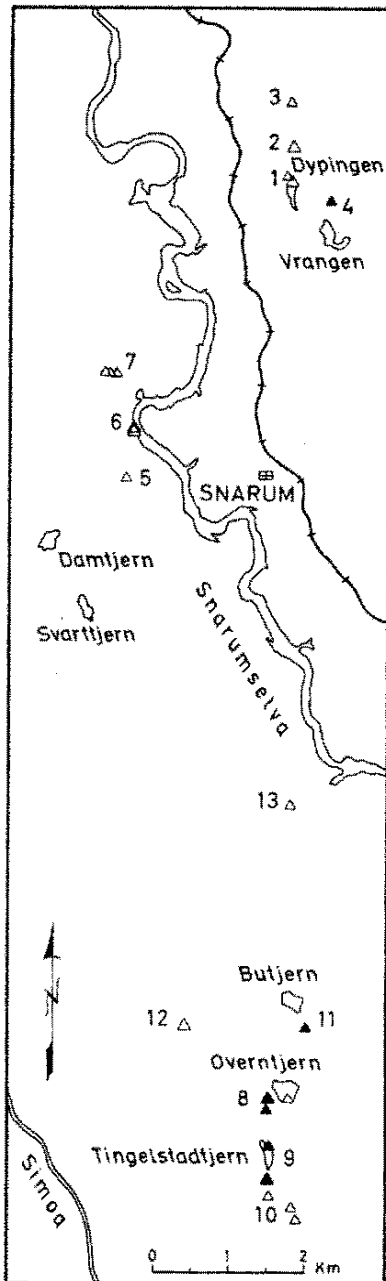
Fig. 1. Simplified map of parts of the Modum area in Buskerud, South Norway. Magnesite occurrences are shown by triangles of two sizes, indicating larger and smaller deposits. The presence of phosphate minerals is denoted by filled symbols. The localities are numbered as follows: (1) Dypingdal (nedre), (2) Dypingdal (øvre), (3) Liaæs, (4) Laupmyr, (5) Nes, (6) Langerud, (7) Sandfall, (8) Overntjern, (9) Tingelstadtjern, (10) Trekasthøgda, (11) Ammundrud, (12) Hulebak, (13) Flaten. From Raade (1986).

stratigraphic horizon. However, ultramafic rocks may well be strata-bound, having intruded preferentially along a weakness zone of a supracrustal sequence. Petrascheck (1971) has tentatively suggested, based on field observations and microscopic studies, that the rocks might have formed from sedimentary magnesites by metasomatic and metamorphic activity. He admits, however, that the evidences are contradictory. Bugge (1978) favoured a magmatic genesis based on the mode of occurrence and the close spatial relations between the magnesite-serpentine rocks and the albitite rocks of the area that he regarded as magmatic differentiates of a deep-seated basaltic magma. In his monograph on the mineralogy of Norway, Neumann (1985, pp. 93 and 130) puts forward the idea of interpreting the magnesite-serpentine rocks as metamorphosed komatiites (olivine-bearing lavas). It is common to all these opinions that they were not based on any geochemical evidence. This is what I wanted to rectify by publishing my previous paper (Raade 1986).

General geochemical characteristics

Magnesium-rich carbonates and silicates are the main phases of the Modum deposits. Forsterite from Dypingdal has the composition $Fo_{98}Fa_2$. Iron is principally present as oxides (hematite and magnetite). Spinel, hydrotalcite and quintinite are the most important aluminium-bearing minerals. Magnesium phosphates and apatite-group minerals occur abundantly only in the Overntjern–Tingelstadtjern area, apatite being otherwise known as a great rarity at Laupmyr and Ammundrud. It should be noted that carbonate is present in holtedahlite and phosphoellenbergerite and is a major constituent of heneuite. Both hydroxylapatite and chlorapatite occur at Tingelstadtjern. Chlorine in amounts up to 0.60 wt.% was recorded by EPMA in hydroxyl-bearing silicates like phlogopite, clinochlore, amesite and pargasite (Raade 1986). It was not noted in serpentine, but is probably present in minor amounts below the EPMA detection limit. Appreciable amounts of titanium occur in hematite and magnetite (cf. Table 2 and Raade 1982) and to a lesser extent in some silicates (phlogopite, clinochlore and pargasite) (Raade 1986). Szaibélyite is a late-formed magnesium borate occurring in veins and along cracks. The two borosilicates kornerupine and tourmaline (species not determined) are confined to the altered outer zone of the Dypingdal deposit. Sulphides (chalcopyrite and pyrite) are extremely rare in all the magnesite-serpentine bodies.

If the hypothesis of a sedimentary dolomitic protolith tentatively forwarded by Jøsang (1966) were true, the calcium originally present must have been almost completely removed. It has been experimentally demonstrated (Johannes 1970) that the formation of magnesite by magnesium metasomatism of primary calcite or dolomite is possible. However, it may be difficult to envisage that a complete exchange can take place within a large rock volume. Metasomatic processes have indeed taken place in the Modum deposits, but to the effect that calcium has been added at a late stage. Small amounts of coexisting dolomite and calcite are either found to replace massive magnesite or occur as late-formed veins associated with brecciation and slickensiding. Carbonate thermometry, based on the Mg content of calcite coexisting with dolomite, gives a temperature of formation for the former case of 520–560°C and ca. 300°C for the latter (Raade 1986). Also the introduction of boron (as szaibélyite) is a late-stage process. A brown apatite mineral rich in thorium is rather abundant in the Overntjern deposit, and a metamict magnesium phosphate with 12–16 wt.% ThO_2 is found in small amounts (Raade 2011). Thorium may have been introduced metasomatically after the formation of phosphates. Spinel and magnetite occurring imbedded in hydrotalcite/quintinite may be considered to be of primary origin because of high contents of nickel and chromium (cf. Table 2 and later discussion). Spinel is extremely low in titanium (Table 2 and Raade 1986), as opposed to hematite and some magnetite. Titanium seems to have been introduced at some later stage in the metamorphic and metasomatic history of the rocks.

Trace elements

Experimental

Trace-element compositions of minerals were determined by INAA at the Mineralogical-Geological Museum (MGM), University of Oslo, using epithermal irradiation. The gamma-ray spectra were recorded with a coaxial Ge(Li) detector and also with a low-energy photon detector (LEPD). A Canberra Series 80 Multichannel Analyzer, Model 8623, was used. The weight of the mineral samples was about 50 mg, and USGS rocks AGV-1 and BCR-1 were applied as standards. Twenty-three samples out of a total of 38 were selected for these analyses. The whole set of 38 samples were at a

later date sent to a commercial laboratory for INAA by thermal irradiation. However, only a limited set of elements can be measured by this method. At the time when the 1986 paper of Raade was prepared, just a small number of data had been processed at the MGM laboratory. Preliminary data for Ni, Co, Sc, Ta and Cs on 22 samples (heneuite was not included) were presented by Raade (1986, Table VII). The full set of trace-element data, from the two laboratories, is listed here in Tables 1 to 4. Not all of these results will be discussed in the present paper; they are collated here in order to be preserved for the future. Minor differences in figures between those in the Raade (1986) table and those given here are the result of using a more refined method of calculation for the latter.

When discussing and interpreting trace-element contents in minerals analysed by INAA, it must be considered that they have been obtained on bulk samples that may contain traces of included phases.
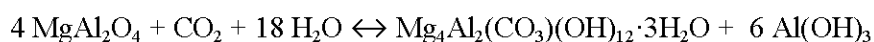
Nickel

Nickel is a key element when it comes to differentiate between sedimentary or magmatic origin of the Modum magnesite-serpentine deposits. The highest contents are found in spinel and magnetite from Dypingdal (304 and 502 ppm Ni, respectively; Table 2), and both are embedded in hydrotalcite. Also the Ni content of hydrotalcite is relatively high, 155 ppm (Table 1). The Ni content of forsterite and serpentine from Dypingdal are 98 and 103 ppm, respectively, and talc, phlogopite and clinochlore have respectively 64, 81 and 144 ppm Ni (Table 3). It should be noted that serpentine from Tingelstadtjern has lower Ni contents of 17 and 32 ppm (Table 3). It is well known that the Ni content of olivine from ultramafic rocks is normally of the order of a few thousand ppm (1000 parts per million equal 0.1%). Faust & Fahey (1962) found a large gap in Ni content between serpentines associated with ultramafic rocks and serpentines from other rock types, with values of around 400-3000 ppm for the former and less than 12 ppm for the latter. The Modum serpentines have values in-between and have definitely not sedimentary affinities. The occurrence of serpentine pseudomorphs after forsterite led Faust & Fahey (1962) to classify the Modum deposits as being of typical ultramafic origin, although they did not measure the actual Ni contents of the Modum samples. The Co values are to some extent positively correlated with the Ni values (Tables 2 and 3). However, the Co content of serpentines is not as good an indicator of their origin as Ni (Faust & Fahey 1962). Relatively high Sc contents are found in oxide minerals (Table 2) and in pargasite from Liaæs (Table 3). The concentration of Sc in serpentine is a rather good indicator of geological origin (Faust & Fahey 1962), and the Modum samples fall in the ultramafic category. In a later contribution by Faust (1963), two samples of serpentine from Modum were analysed for Ni (51 and 78 ppm), Cr (2 and 9 ppm), Co (<2 and 8 ppm) and Sc (6 and 2 ppm), confirming their ultramafic affinity.

Trace elements in oxides and hydrotalcite/quintinite

Relatively high concentrations of the trace elements Co, Cr, Ni, Zr and Hf occur in spinel and magnetite from Dypingdal (Table 2). Hydrotalcite/quintinite that surrounds these minerals seems to have inherited this trace-element pattern (Table 1). The contents of Co, Cr and Ni are too high to be entirely caused by inclusions of spinel, and these elements can easily enter the hydrotalcite structure. The formation of hydrotalcite ($Mg_6Al_2(CO_3)(OH)_{16} \cdot 4H_2O$) and quintinite ($Mg_4Al_2(CO_3)(OH)_{12} \cdot 3H_2O$) must have taken place by reaction of CO_2-bearing fluids with primary spinel according to the following principal reactions. Either diaspore or gibbsite may form in the process, and both have been

observed as intergrowths with hydrotalcite/quintinite. The equilibrium mole fraction of CO_2 in the fluid phase for these reactions is low, varying from 0.036 to 0.077.

$$6 \, MgAl_2O_4 + CO_2 + 17 \, H_2O \leftrightarrow Mg_6Al_2(CO_3)(OH)_{16} \cdot 4H_2O + 10 \, AlO(OH) \text{ (diaspore)}$$

$$4 \, MgAl_2O_4 + CO_2 + 12 \, H_2O \leftrightarrow Mg_4Al_2(CO_3)(OH)_{12} \cdot 3H_2O + 6 \, AlO(OH)$$

$$6 \, MgAl_2O_4 + CO_2 + 27 \, H_2O \leftrightarrow Mg_6Al_2(CO_3)(OH)_{16} \cdot 4H_2O + 10 \, Al(OH)_3 \text{ (gibbsite)}$$

$$4 \, MgAl_2O_4 + CO_2 + 18 \, H_2O \leftrightarrow Mg_4Al_2(CO_3)(OH)_{12} \cdot 3H_2O + 6 \, Al(OH)_3$$

Minor and trace elements in magnesite

A review of several minor and trace elements in magnesite was published by Möller (1989), including *inter alia* Ni, Co and Cr. It was stressed that results on bulk samples may be unreliable with regard to mineralogical purity of the samples. The data were found to be of little relevance when compared to the results on Modum magnesites given in Table 1. Modum magnesites are from ophimagnesite or from large cleavage fragments and are thought to be quite pure. Only 8 ppm Cr was detected in one of the ophimagnesite samples, and Ni is 13 and 19 ppm in two such samples. Also Co is low with a maximum of 10–12 ppm in magnesite cleavage fragments. This is in contrast to Cr values of 100 to 4000 ppm, Ni from 10 to 100 ppm and Co from 50 to 150 ppm for a limited number of coarse-grained (sparry) magnesites thought to be formed by CO_2 metasomatism of ultramafic rocks (Möller 1989). Magnesites from one such occurrence, the Ochsner serpentinite in Austria, have Cr in excess of 4000 ppm (Dulski & Morteani 1989). Evidently, such amounts of Cr^{3+} can hardly enter the magnesite crystal lattice.

Coarsely crystalline magnesite from Modum has 0.06–0.13 wt.% CaO, whereas ophimagnesite has 0.12–0.20 wt.% CaO; the corresponding FeO values are 0.84–1.26 and 0.24–0.52 wt.% (Raade 1986). These differences are not easily explained. Ophimagnesite has 33 and 114 ppm Sr, but only 3 ppm were found in coarse magnesite (Table 1). Ba is not included in the table as only two determinations were made; these are 34 and 20 ppm for samples 9 and 10, respectively. Also Rb is not included; three determinations gave 2.1, 1.5 and 2.1 ppm for samples 3, 10 and 11, respectively. Most ultramafic rocks have very low contents of Sr, whereas metagabbros of the Modum district have up to 320 ppm Sr (Munz *et al.* 1994). Ba values up to 70 ppm have been recorded from ultramafic rocks (Puchelt 1972). Rare earth elements are very low in the Modum carbonates. Because of erratic results, REE data are not included in Table 1. However, sample 10, a coarse magnesite from Tingelstadtjern, has (in ppm): La 0.9, Ce 3, Sm 0.05, Eu 0.04, Tb 0.02, Tm 0.04, Yb 0.74 and Lu 0.15. These results are given just for the record and will not be discussed.

Arsenic

Relatively low and variable contents of arsenic are found in carbonates and oxides (Tables 1 and 2), and higher contents occur in some of the phyllosilicates (Table 3). Much higher concentrations are present in the phosphate minerals (Table 4). Raadeite contains on average 0.40 wt.% As_2O_5, and the associated minerals holtedahlite, althausite and apatite have 0.08–0.31, 0.17 and 0.68 wt.% As_2O_5, respectively (Chopin *et al.* 2001). EPMA analysis of heneuite gave 1.05 wt.% As_2O_5 (Raade *et al.* 1986), to be compared with 0.86 and 0.95 wt.% As_2O_5 as recalculated from ppm As values in Table 4. Phosphoellenbergerite has as much as 5.5 wt.% As_2O_5 (Raade *et al.* 1998).

Limestones and dolomites generally have low As contents with average values around 1–2 ppm (Onishi & Sandell 1955a, Onishi 1969). On the other hand, phosphorites and other sedimentary phosphate rocks typically have averages around 20 ppm As (Onishi 1969, Nathan 1984). For peridotites, dunites, serpentinites and gabbros, As contents are rarely above 6 ppm (Onishi & Sandell 1955a). A carbonate-sedimentary precursor of the Modum magnesite deposits can be ruled out and magmatic-metasomatic processes introducing P and As seem to be likely.

Antimony

Contents of Sb in excess of 6 ppm are present in hematite, and around 2 ppm are found in magnetite (Table 2). Magnesites from ophimagnesite have 0.23 and 0.78 ppm Sb (Table 1). The Sb content of serpentine may reach 1 ppm although it was not found in forsterite (Table 3). Concentrations of Sb and As are not correlated in the Modum minerals. For comparison, contents up to 0.11 ppm As are reported in titaniferous magnetite and up to 0.27 ppm in ilmenite from the Skaergaard intrusion, Greenland, and iron-rich olivine may have up to 1.43 ppm (Esson *et al.* 1965). Average values of Sb in ultramafic rocks and limestones are 0.1 and 0.3 ppm, respectively (Onishi & Sandell 1955b). The Sb contents of the Modum oxide minerals are surprisingly high.

Chemistry of apatites, including rare earth elements

EPMA analytical data for two apatite minerals from Tingelstadtjern, combined with CO_2 and H_2O determinations performed with a Perkin-Elmer elemental analyser, were published by Raade (1986). A light grey apatite is chlorine-dominated with $(Cl_{0.57}F_{0.32}OH_{0.22})_{\Sigma1.11}$, based on 3 (P+C) apfu (atoms per formula unit). The CO_2 content is 1.07 wt.%. A darker grey apatite is slightly OH-dominated with $(OH_{0.32}F_{0.28}Cl_{0.13})_{\Sigma0.73}$, also based on 3 (P+C) apfu. The reason for the deficiency at the halogen/OH site in this apatite is not known; it could be the effect of an oxy component. The CO_2 content is 0.70 wt.%. These apatites coexist with althausite and the other magnesium phosphate minerals mentioned in the Introduction. A dark brown, Th-bearing apatite occurs at Overntjern. Only an incomplete EPMA analysis was made for this mineral, without quantification of the ThO_2 content. Althausite is abundant also at the Overntjern locality.

Apatite compositions from mafic complexes and lithospheric mantle veins show that fluids with a mantle provenance may be enriched in chlorine relative to fluorine and hydroxyl (Piccoli & Candela 2002). This gives credit to the idea that the Modum apatites are derived from a mafic or ultramafic mantle source.

Contents of REEs (rare earth elements) for all three apatites were determined by INAA. The chondrite-normalized REE patterns are displayed in Raade (1986). All of them show a negative Eu anomaly. The Th-rich apatite from Overntjern has a remarkably different pattern compared to the other two and shows strong depletion of La and Ce and a distinct increase in Lu. Parekh & Möller (1977) have developed a discrimination diagram involving Yb and La, which seems to differentiate between apatites of magmatic and sedimentary origin. The Modum samples plot in the magmatic field.

Comparison with apatites from the well-known apatite vein deposits at Ødegårdens verk in the Bamble Sector, genetically connected to gabbroic intrusions, reveals no immediate chemical relationships to the Modum apatites. Chlorapatite with veins of secondarily formed hydroxylapatite occur at Øde-

gården. These are devoid of fluorine, very low in carbonate (0.08–0.09 wt.% CO_2), and arsenate was not detected (Morton & Catanzaro 1964). Little is known about the distribution of carbonate-rich apatite ("dahllite") in these deposits. It is interesting to note that the magnesium phosphate mineral wagnerite, $Mg_2(PO_4)F$, has been reported from several places at Ødegården and in the adjacent Havredal locality (Neumann 1985).

Stable O and C isotopes in carbonates

Data on oxygen and carbon isotope compositions of five coarsely crystalline (sparry) magnesite samples, two ophimagnesite samples and one sample of coexisting dolomite and calcite from a fissure-filling in serpentine are listed in Table 5. The low $\delta^{18}O$ value of 4.4 ‰ for the latter is distinctly different from $\delta^{18}O$ values of the other samples which are remarkably similar and vary from 11.1 to 13.4 ‰. The two ophimagnesite samples are slightly enriched in the heavy carbon isotope ($\delta^{13}C$ = – 1.3 and –1.6 ‰) compared to the sparry magnesites. Two different specimens of magnesite cleavage fragments from Dypingdal gave very close values for $\delta^{13}C$ and $\delta^{18}O$, indicating a high degree of isotopic homogeneity.

Raade (1986) has plotted $\delta^{18}O$ (SMOW) versus $\delta^{13}C$ (PDB) for the Modum samples and added literature data available at that time on magnesites from five worldwide localities. Those considered to be of sedimentary origin are characterized by positive $\delta^{13}C$ values and high $\delta^{18}O$ values (up to 30 ‰). This is clearly at variance with data for the Modum magnesites.

A large number of carbon and oxygen isotope data on magnesites was collated by Kralik et al. (1989). Two samples of Modum magnesite show $\delta^{18}O$ 9.8 and 9.9 ‰ and $\delta^{13}C$ 0.7 and 1.4 ‰; for two other samples the values are $\delta^{18}O$ 11.5 and 14.2 ‰ and $\delta^{13}C$ –4.4 and –0.5 ‰ (three of the data sets are from Zachmann 1989). The type of magnesite is not recorded but they are probably ophimagnesites. Kralik et al. (1989) concluded that cryptocrystalline to fine-grained magnesite in fractures of ultramafic rocks has a meteoric carbon source and that "most coarse grained spar magnesites crystallized or recrystallized from still disputed precursor carbonates during metamorphic or hydrothermal events".

Raade (1986) noted that the Modum samples plot in the same area as carbonatites (Deines & Gold 1973) and concluded that the evidence seems to favour metasomatism with CO_2 of mantle-derived origin. A sedimentary origin of the Modum magnesite deposits was considered highly unlikely. It is significant that measurements of magmatogenic CO_2 exhalations yield $\delta^{13}C$ values between –2 and –6 ‰ (Hoefs 1973, Allard 1979), cf. Modum magnesite data in Table 5. Regional availability of CO_2-bearing fluids in the Modum area is testified by extensive hydrothermal calcite deposits at Gampehue in Sigdal and a persistent content of calcite in albitite rocks (Munz et al. 1994). However, a genetic relationship between these calcite occurrences and the magnesite deposits is not likely. Munz et al. (1994) concluded, based on age relationships and Nd and Sr isotopic signatures, that the albite-rich rocks did not form from either melts or fluids cogenetic with the gabbros and suggested a crustal source of the albitizing fluids. Stable isotopic data seem not to be available for these calcites.

An occurrence of coarsely crystalline dolomite in large amounts as veins and tabular bodies in association with albitites within the Late Precambrian greenschist-facies metamorphics of Sinai was described by Bogoch et al. (1986). The dolomite has mantle-isotopic characteristics ($\delta^{18}O$ 5.3 to 8.5 ‰ and $\delta^{13}C$ –7.5 to –8.4 ‰) and high contents of Cr (77 ppm), Ni (21 ppm) and Sc (70 ppm) which is

different from carbonatites. Taking into consideration also Sr and Nd isotopic data, a multistage model based on a carbonate-bearing peridotitic precursor was suggested (Bogoch *et al.* 1986).

Discussion

Comparison with some mineralogically similar carbonate rocks

It is well known that minerals like forsterite (with alteration to serpentine), diopsidic clinopyroxene, phlogopite and spinel may form in "impure", siliceous limestones and dolomites that have been subjected to regional metamorphism. Such rocks are for instance known from the high-grade metamorphic Precambrian of Rogaland (Sauter 1981) and from the Caledonian migmatite and gneiss area of NW Spitsbergen (Bucher-Nurminen 1981).

The Lona mineral locality at Sannidal (Aust-Agder), protected by law against collecting, has some mineral associations that resemble those at Modum. These are the occurrence of blue spinel mantled by hydrotalcite and forsterite pseudomorphosed to serpentine. The Modum area belongs to the Precambrian Kongsberg Sector, and the Lona occurrence is situated in the Bamble Sector. The two sectors are separated by the Permian Oslo graben and are assumed to represent a former continuous area. I sampled the Lona rocks and minerals in 1981 but unfortunately did not have the time to investigate the material in any detail. Additional identified minerals are coexisting calcite and dolomite, late-formed aragonite, diopside, tremolite, phlogopite, unspecified apatite, graphite and secondary rozenite formed on pyrrhotite. Based on mode of occurrence, as a layer in a supracrustal sequence, and mineral associations, the Lona carbonate rock is believed to have a sedimentary origin.

The name "houghite" was introduced by Shepard (1851) for a mineral that later was shown to be identical with hydrotalcite (Frondel 1941). It was found at Somerville, Rossie, St. Lawrence County, New York. It is interesting to note that it forms pseudomorphs after spinel, thus resembling the spinel–hydrotalcite associations found at Dypingdal and at Lona. Associated minerals of "houghite" are dolomite, scapolite, phlogopite, graphite and serpentine (Johnson 1851). The minerals occur in a rock described as crystalline limestone, occurring near beds of sandstone (Johnson 1851). The primary rock was no doubt a calcareous sediment that was subjected to metasomatic and metamorphic activity.

Although the Modum magnesite rocks show mineralogical similarities to the above examples, their origin must be quite different, as argued elsewhere in this paper. A significant contrast is the occurrence of enstatite instead of diopside at Modum which shows that the initial mineral assemblage was extremely poor in calcium.

Origin of magnesite rocks

Number 28 of *Monograph Series on Mineral Deposits* (1989), edited by Peter Möller, was devoted entirely to magnesite, but my 1986 paper is not mentioned. Some individual chapters are referred to here. It gives the overall impression that the origin of some magnesite rocks has been, and still is, quite controversial. Different origins have been proposed in the past for single deposits to the extent that mineralizing fluids may have been either descending or ascending (Pohl 1989). Pohl & Siegl (1986) concluded that "the interpretations of $\delta^{18}O$ values offered by different authors appear quite contradictory". Magnesite $\delta^{18}O$ values of 7 to 8.5 ‰ and $\delta^{13}C$ of –3.5 to –4 ‰ were reported from the Ochsner deposit in Austria (Dulski & Morteani 1989), regarded to be formed by CO_2 metasomatism of

ultramafic rocks. The authors noticed that these values fall within the ranges typical for high-grade metamorphic rocks and are also near to carbonatite isotope values. Their conclusion was that the $\delta^{18}O$ data are consistent with a fluid phase of mainly meteoric water at a temperature of about 550°C, but the $\delta^{13}C$ data were not considered in their interpretation. It should be noted that these C and O isotope values are in the vicinity of, but distinct from, data on Modum magnesites.

As an overall summary, most magnesite deposits are assumed to be of sedimentary origin. Those associated with ultramafic rocks can form by weathering or hydrothermal alteration of such rocks, resulting in fine-grained or microcrystalline veins and stockworks of magnesite. Extensive CO_2 metasomatism of ultramafic rocks involves replacement reactions in situ and results in sparry magnesite. Subsequent metamorphic episodes make the picture more complicated.

The magmatic-metasomatic rock type sagvandite deserves to be mentioned briefly, although not related to the Modum magnesite deposits. It was originally described by Pettersen (1883) from Balsfjord, Troms, and is a carbonate orthopyroxenite consisting of about 10% ferrous magnesite and 90% bronzite (Barth 1926). Such rocks, also with olivine, have since been described from many places, and there exists a comprehensive literature on the subject. As a curiosity, Zachmann (1989, Table 4) assigned the enstatite-bearing Modum (Snarum) magnesite rocks to sagvandite.

Conclusions

The complex metamorphic and metasomatic history of the Modum magnesite deposits is not easily unravelled. It seems clear that forsteritic olivine, enstatite and spinel were constituent phases of a primary ultramafic rock. Enstatite has been identified from the Dypingdal, Overntjern and Tingelstadtjern deposits. It is mainly found in the peripheral parts of the magnesite-serpentine bodies and is always more or less altered to talc. Remnants of enstatite in talc from Dypingdal has a high Al_2O_3 content of 2.89 wt.%, indicating a minimum temperature of formation of about 760°C, whereas two relatively fresh enstatites from Overntjern have 0.92 and 0.47 wt.% Al_2O_3, corresponding to minimum temperatures of ca. 540–470°C (Raade 1986, based on Gasparik & Lindsley 1980). Metamorphic conditions in the Modum area are given as 600–750°C and 6–8 kbar by Munz (1990). The gabbroic rocks of the area intruded at 1000–1100°C and 5–8 kbar and equilibrated at 600–800°C and 7–10 kbar (Munz & Morvik 1991).

Serpentinization of the ultramafic bodies at Modum may have preceded CO_2 metasomatism or occurred concurrently according to the reaction: 2 forsterite + 3 H_2O ↔ 1 serpentine + 1 brucite (Johannes 1968). Initially formed brucite reacts with CO_2 to form magnesite: $Mg(OH)_2$ + CO_2 ↔ $MgCO_3$ + H_2O. The chlorine content of the rocks may have played an active role in the breakdown of olivine during serpentinization, whereby Fe^{2+} in olivine is taken into solution via an intermediate Cl-bearing phase (Rucklidge & Patterson 1977, Miura et al. 1981). Iron is thus redistributed and normally separates as magnetite under relatively reducing conditions (Moody 1976). Hematite is the prevailing iron oxide in the Modum deposits, requiring more oxidizing conditions. As CO_2 metasomatism obviously has taken place, olivine may have reacted directly to serpentine plus magnesite according to the reaction: 2 forsterite + 2 H_2O + CO_2 ↔ 1 serpentine + 1 magnesite (Johannes 1969). The reaction of CO_2-bearing fluids on spinel, producing hydrotalcite/quintinite plus diaspore/gibbsite, is discussed above.

Main elements of primary silicates and oxides in the Modum deposits are Mg, Fe and Al. The trace-element contents of these minerals must be primordial as well: Ni, Co, Cr, Sc, Zr, Hf, Ta, Sb and As

(cf. Tables 2 and 3). Late-introduced elements are C, B, Ca and Th, whereas Ti could be both primary and late. The source of Sr, Rb, Ba and Cs is uncertain. Locally important concentrations of Mg and Ca phosphates with As and REEs, Cl and F, must have a magmatic origin. Although levels of Ni and Cr are below those expected for an ultramafic precursor, they are certainly higher than for sedimentary-derived magnesite deposits, as discussed above. It is possible that Ni was largely removed by sulphide segregation at depth; the Modum magnesite deposits are conspicuously poor in sulphides, and large-scale Ni-bearing sulphide deposits related to gabbroic intrusions are known in the Ringerike district north-east of Modum. The mineralogical and geochemical characteristics referred to here, coupled with results on O and C isotopes in magnesite, leave no doubt that the Modum magnesite deposits are of ultramafic origin, having been subjected to metasomatic introduction of CO_2 from a mantle source.

A note from the author

It seems appropriate to point out that the present paper builds heavily on my 1986 paper, and that no systematic literature search has been made in later years, although several new references have been added here. This may not necessarily detract from the conclusions presented.

Acknowledgements

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Table 1. *Neutron activation analytical data for carbonates from Modum magnesite deposits (ppm).*

		1	2	3	4	5	6	7
		Magnesite	**Magnesite**	**Dolomite**	**Magnesite**	**Magnesite**	**Magnesite**	**Magnesite**
		Overntjern	*Dypingdal*	*Dypingdal*	*Langerud*	*Overntjern*	*Langerud*	*Dypingdal*
				EPMA M4	EPMA M5			
		Cleavage	Cleavage	With calcite	Ophimagnesite	Cleavage	Cleavage	Ophimagnesite
As	GR	–	–	0	–	–	–	–
As	Ext lab	0	1.8	2.3	2.1	0	0	0
Co	GR	–	–	6.2	–	–	–	–
Co	Ext lab	12	10	8	6	4	9	8
Cr	Ext lab	0	0	0	0	0	0	0
Cs	GR	–	–	0	–	–	–	–
Ni	GR	–	–	2	–	–	–	–
Sb	GR	–	–	0	–	–	–	–
Sb	Ext lab	0	0	0	0	0	0	0.3
Sc	GR	–	–	1.3	–	–	–	–
Sc	Ext lab	2.1	2.6	1.2	4.8	4.2	9.8	1.9
Sr	GR	–	–	148	–	–	–	–
Ta	GR	–	–	2.52	–	–	–	–
U	GR	–	–	0.3	–	–	–	–

		8	9	10	11	12	13	14
		Magnesite	**Magnesite**	**Magnesite**	**Magnesite**	**Dolomite**	**Hydrotalcite**	**Hydrotalcite**
		Dypingdal	*Nes*	*Tingelstadtjern*	*Tingelstadtjern*	*Tingelstadtjern*	*Dypingdal*	*Dypingdal*
		EPMA M6				EPMA M7		
		Cleavage	Ophimagnesite	Cleavage	Ophimagnesite	With calcite	White	Pale blue
As	GR	–	2.5	0.6	17.8	4.7	0.7	–
As	Ext lab	0	1.6	0	12.4	3.1	0	0
Co	GR	–	1.7	1.7	1.7	1.4	5.8	–
Co	Ext lab	6	2	6	3	4	9	10
Cr	Ext lab	0	0	0	8	0	50	46
Cs	GR	–	0	0.14	0.16	0.61	0	–
Ni	GR	–	19	5	13	7	155	–
Sb	GR	–	0.23	0.05	0.78	0.04	0.08	–
Sb	Ext lab	0	0	0	0.8	0	0	0
Sc	GR	–	3.2	5.9	3.6	2.4	0.4	–
Sc	Ext lab	6.1	3.0	6.1	3.5	2.4	0.3	0.4
Sr	GR	–	33	3	114	120	119	–
Ta	GR	–	0.02	0.01	0.04	0.01	0.01	–
U	GR	–	0.3	0.2	0.4	0.3	0.3	–
Hf	GR	–					0.9	–
Zr	GR	–					43	–

Explanations to Tables 1 – 4:

"–" means not analysed   "0" means not detected (below detection limit)

Blank fields: unreliable results or below detection limit

Underlined numbers are averages of two to six single determinations

GR: Analyses performed by G. Raade at the Mineralogical-Geological Museum 1984 (epithermal irradiation)

Ext lab: Analyses performed by an external, commercial laboratory 2002 (thermal irradiation)

Table 2. *Neutron activation analytical data for oxides from Modum magnesite deposits (ppm).*

		15	16	17	18	19	20
		Spinel	**Spinel**	**Magnetite**	**Magnetite**	**Hematite**	**Hematite**
		Dypingdal	*Dypingdal*	*Dypingdal*	*Hulebak*	*Dypingdal*	*Overntjern*
		Separate	Separate		EPMA M22	EPMA old 1	EPMA M18
		Nonmagnetic	Magnetic	In hydrotalcite		Crystal	Twinned
As	GR	3	–	84	19	0	16
As	Ext lab	0	0	8.8	6.8	3.5	7.6
Co	GR	<u>48</u>	–	<u>161</u>	<u>29</u>	<u>3</u>	<u>1</u>
Co	Ext lab	60	65	177	30	12	5
Cr	Ext lab	248	215	55	75	33	99
Hf	GR	<u>21.1</u>	–	<u>4.0</u>	<u>5.8</u>	<u>9.6</u>	~ <u>0.5</u>
Hf	Ext lab	17	1	0	0	7	0
Ni	GR	304	–	502	19	16	0
Sb	GR	<u>0.41</u>	–	<u>1.90</u>	<u>1.25</u>	<u>6.43</u>	<u>6.32</u>
Sb	Ext lab	0.4	0	1.8	2.7	6.1	6.6
Sc	GR	<u>1.0</u>	–	<u>19.5</u>	<u>88.4</u>	<u>46.9</u>	<u>35.0</u>
Sc	Ext lab	1.3	2.7	19	100	46.7	39.9
Sr	GR	297	–	349	50	3	34
Ta	GR	<u>0.09</u>	–	<u>1.36</u>	<u>5.59</u>	<u>2.85</u>	<u>2.11</u>
Zr	GR	<u>719</u>	–	143	<u>120</u>	<u>216</u>	6
U	GR	0.4	–	<u>0.2</u>	0.3	<u>0.4</u>	0.4
TiO₂ %	GR	0.01	–	2.16	6.67	1.86	1.33

Comments on individual elements in Tables 1 – 4:

As: There are discrepancies between GR and Ext lab results; the short half-life of the product isotope [76]As (26.3 h) should be considered. Epithermal irradiation is preferred, and the GR data are deemed to be best. **Ba**: Thermal irradiation is not suited. **Co**: Reasonable agreement between GR and Ext lab results. **Cr**: Thermal irradiation is preferred. **Cs**: Only by epithermal irradiation. **Hf**: Good agreement between GR and Ext lab results; better sensitivity with epithermal irradiation. **Ni**: Only by epithermal irradiation. **Rb**: Epithermal irradiation preferred. **Sb**: Reasonable agreement between GR and Ext Lab results; better sensitivity with epithermal irradiation. **Sc**: Excellent agreement between GR and Ext lab results. **Sr**: Only by epithermal irradiation. **Ta**: Only by epithermal irradiation. The 1121.2 keV peak from [182]Ta interferes with [46]Sc at 1120.5 keV and was not used. **Th**: Epithermal irradiation is preferred. **Ti**: Excellent agreement with EPMA values for hematite and magnetite reported by Raade (1986). TiO₂ is 0.92 wt.% for sample 34 (pargasite) by EPMA (Raade 1986) versus 0.97 wt.% by INAA (Table 3). **Zr**: Both thermal and epithermal irradiation can be used, but it was not analysed by Ext lab. **U**: Epithermal irradiation is preferred. **REEs**: Nine rare earth elements were analysed; for eight of them, both thermal and epithermal irradiation can be used. The exception is Lu which requires thermal irradiation. There is generally a good agreement between GR and Ext lab results.

Table 3. *Neutron activation analytical data for silicates from Modum magnesite deposists (ppm).*

		21	22	23	24	25	26	27
		Forsterite	Talc	Talc	Phlogopite	Clinochlore	Serpentine	Serpentine
		Dypingdal	*Overntjern*	*Dypingdal*	*Dypingdal*	*Dypingdal*	*Overntjern*	*Overntjern*
		XRD 1759		XRD 1695	EPMA M12	EPMA M13	XRD 21828	
		Separated	Platy	Massive			Chrysotile	Hematite incl.
As	GR	152	399	2.5	5	56	–	–
As	Ext lab	25.9	55.7	0	0	9.1	67.4	80
Ba	GR				293			
Co	GR	18	2	15	5	20	–	–
Co	Ext lab	20	5	16	11	23	11	4
Cr	Ext lab	0	0	0	32	11	0	0
Cs	GR	0	0.06	0.13	3.2	0.06	–	–
Ni	GR	98	42	64	81	144		
Rb	GR				367			
Rb	Ext lab				233			
Sb	GR	0	0.19	0.25	0.30	0.21	–	–
Sb	Ext lab	0	0.3	0.2	0.8	0	0	1.7
Sc	GR	7.1	0.8	3.5	14.4	10.0	–	–
Sc	Ext lab	7.4	0.6	4.2	12.7	10.6	0.6	3.3
Sr	GR	100	42	58	71	112	–	–
Ta	GR	0.01	0.01	0.02	1.18	0.36	–	–
U	GR	0.3	0.5	0.5	0.2	0.3	–	–

		28	29	30	31	32	33	34
		Serpentine	Serpentine	Serpentine	Serpentine	Enstatite	Enstatite	Pargasite
		Tingelstadtjern	*Tingelstadtjern*	*Dypingdal*	*Hulebak*	*Overntjern*	*Tingelstadtjern*	*Liaes*
								EPMA 3/5/82
		Yellow-green	Darker green	Pseudomorph	Yellow-green	Hematite incl.	Hematite incl.	
As	GR	762	384	101	–	59	–	57
As	Ext lab	126	65.6	14.6	10.9	12	29.3	9.9
Co	GR	1.5	3	8	–	2	–	10
Co	Ext lab	6	4	10	42	13	4	16
Cr	Ext lab	0	0	0	0	0	0	0
Cs	GR	0.02	0.12	0.06	–	2.5	–	1.7
Ni	GR	17	32	103	–	9	–	30
Sb	GR	0.98	1.09	0.32	–	0.55	–	0.33
Sb	Ext lab	0	0.9	0.6	0	0.3	0	0
Sc	GR	3.8	3.8	6.6	–	4.8	–	117
Sc	Ext lab	4.0	3.9	7.6	5.6	4.9	13.7	130
Sr	GR	30	32	96	–	0	–	29
Ta	GR	0.04	0.06	0.06	–	0.04	–	1.55
U	GR	23.9	0.4	0.2	–	0.4	–	0.2
Hf	GR / Ext							8.6 / 8
Zr	GR							470
Th	GR / Ext							3.7 / 3.9
La	GR / Ext							6.0 / 7.2
Ce	GR / Ext							39 / 41
Nd	Ext lab							38
Sm	Ext lab							17
Eu	GR / Ext							3.7 / 4.1
Tb	GR / Ext							4.3 / 4.3
Tm	GR							3.0
Yb	GR / Ext							25.1 / 25.7
Lu	Ext lab							3.75
TiO_2 %	GR							0.97

Table 4. *Neutron activation analytical data for phosphates from Modum magnesite deposits (ppm).*

		35 **Althausite** *Overntjern* Brownish	36 **Althausite** *Tingelstadtjern* Grey	37 **Heneuite** *Tingelstadtjern* Type material	38 **Apatite** *Tingelstadtjern*
As	GR	–	7048	5590	–
As	Ext lab	8400	9020	6200	705
Ni	GR	–	3	4	–
Sc	GR	–	20.5	71	–
Sc	Ext lab	123	24.7	83.2	1.3
Sr	GR	–	345	56	–
Th	GR	–	0.18	2.3	–
Th	Ext lab	0	2.2	0	18.4
La	GR	–	9.7	27.4	–
La	Ext lab	0	5.0	28.3	500
Ce	GR	–	8	64	–
Ce	Ext lab	0	10	102	1740
Nd	Ext lab	0	0	100	1200
Sm	GR	–	0	25.5	–
Sm	Ext lab	0.3	2.6	32.2	286
Eu	GR	–	0.7	7.2	–
Eu	Ext lab	0	0	9.7	80.3
Tb	GR	–	0.3	6.1	–
Tb	Ext lab	0	0	7.5	60
Tm	GR	–	0.1	2.2	–
Yb	GR	–	1.2	16.3	–
Yb	Ext lab	0	0.8	19	288
Lu	Ext lab	0	0.12	3.0	41.3

Table 5. *Oxygen and carbon isotope compositions of carbonates from Modum (per mille).*

No.	Mineral	Type	Locality	δ13C (PDB) ‰	δ18O (SMOW) ‰
1	Magnesite	Coarse cleavages	Overntjern	– 8.0	11.4
2	Magnesite	Coarse cleavages	Dypingdal	– 4.9	11.6
3	Dolomite + calcite	Fissure filling	Dypingdal	– 4.2	4.4
4	Magnesite	Ophimagnesite	Langerud	– 1.3	13.4
5	Magnesite	Coarse cleavages	Overntjern	– 9.0	12.4
6	Magnesite	Coarse cleavages	Langerud	– 3.6	11.6
7	Magnesite	Ophimagnesite	Dypingdal	– 1.6	12.6
8	Magnesite	Coarse cleavages	Dypingdal	– 5.2	11.1