

# Industrial mineral quality of limestone: the effect of contact metamorphism on textural properties, brightness and geochemistry

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When carbonates are altered by contact metamorphism the industrial mineral quality of the rock can be affected. Based on two contrasting limestone units from the Oslo Rift, Norway, a study has been made of the impact that contact metamorphism has on the quality of limestone with respect to its textural properties, brightness and geochemistry. Samples with variable metamorphic imprints have been examined and compared to their non-metamorphosed equivalents. The results indicate that, in the context of limestone utilisation for industrial mineral applications, contact metamorphism can have a favourable effect on some properties. It can reduce the content of organic matter and give the rock a lighter colour and increased brightness values, and the calcite can attain more even grain-shape and -boundaries and increased grain size. However, there are no indications from the present study that contact metamorphism improves the quality with respect to the occurrence and abundance of contaminants. These limestones show that the impurities can remain very fine-grained and partly intergrown with the calcite, implying that contact-metamorphosed limestone may be no simpler to purify than non-metamorphosed limestone. If the marble has also been metasomatically altered, which is not uncommon in contact-metamorphic environments, the rock is actually likely to be more contaminated, and of correspondingly poorer quality, than it was originally. Overall, the implications are that contact metamorphism need not improve the limestone quality sufficiently to produce an economic industrial mineral deposit. It may be required that the limestone was of very high purity prior to metamorphism and also has been unaffected by any accompanying metasomatism.

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## Introduction

Carbonate rocks, especially limestone but also dolostones, are very important industrial mineral resources worldwide, both in terms of production volume and market value. They have a large number of uses. The various end uses have very diverse quality requirements with regard to both chemical and physical properties of the rock. Whether a carbonate rock can be used and the quality and price which can be obtained for the industrial mineral product is, to a large degree, dependent on the amount and occurrence of impurities in the rock. Almost monomineralic limestone containing approximately 100% calcite is rare; in most deposits, lesser or greater amounts of contaminating components are present. Common mineral inclusions are quartz, calc-silicates, graphite, and sulphides. Textural properties such as grain size, shape and boundaries, the occurrence of contaminants as single grains, exsolutions, or in the crystal lattice, the nature of intergrowths, etc., are critical factors which determine whether the impurities can be liberated by grinding and removed during the beneficiation process.

The purest deposits, which represent raw materials for fine-grade, ultra white fillers, are commonly localised in areas of high metamorphic grade. It can thus be expected that metamorphism is an important mechanism which contributes to the formation of carbonate qualities that are suitable for exploitation. One common and well-known effect of

metamorphism is an increase in the rock's grain size. Another favourable effect, commonly observed in limestone units but less well documented, is bleaching of the rock colour.

For industrial mineral purposes, it is of interest to improve our understanding and documentation of the impacts that metamorphism has on the properties of carbonate rocks. This paper presents the results of a study undertaken in this context. The investigation was restricted to contact metamorphism and was aimed at identifying what type of effects this geological process has on the industrial mineral quality of limestone with respect to its textural properties, brightness and geochemistry. More detailed descriptions of the investigations have been presented in Kjølle (1998).

## Geology and sampling

The limestone units selected for the study are located in the Oslo Rift region of southern Norway. The Oslo Rift was a natural choice because it is one of few regions in Norway almost unaffected by regional metamorphism. The primary features are well preserved in the limestones and can be compared to the metamorphic effects in the contact-metamorphosed limestone commonly found adjacent to the many intrusions of this region.

Limestone of original high purity was preferred for the study. In the Oslo Rift, such units are uncommon, but a few relatively pure limestones occur. All limestones of the Oslo

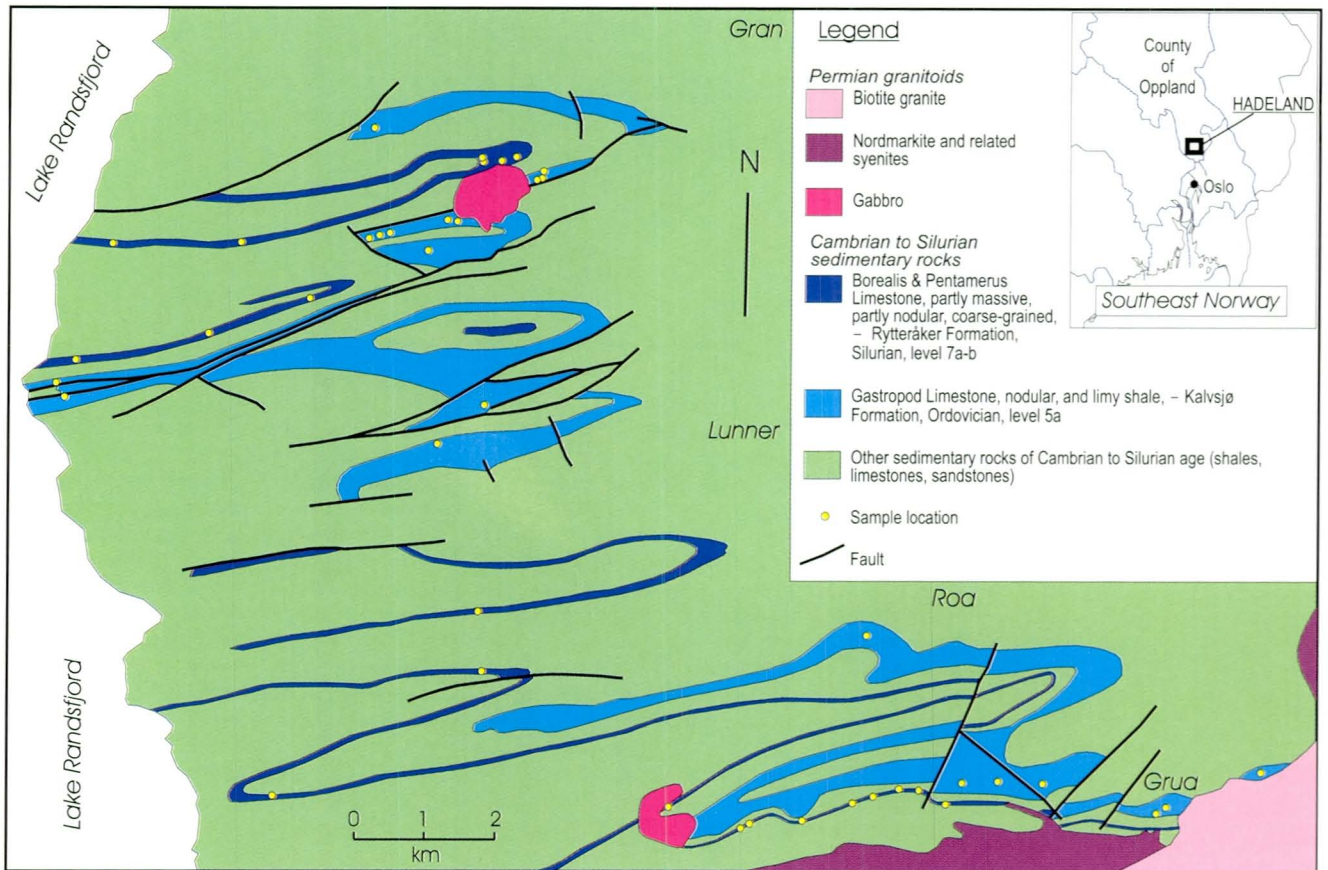


Fig. 1. Geology of the Hadeland area with sample locations. The map shows a simplified geology and is based on the preliminary bedrock map Gran 1815-1, 1:50,000 (Olerud & Owen 1995).

Rift are, to some extent, contaminated by shale, usually occurring in the form of nodular limestone beds which rapidly alternate with thinner shale layers. Limestone formations sampled were selected based on criteria of relatively high purity, availability for sampling in non-metamorphosed and contact metamorphic form, with sufficient exposure along strike, and of massive character or as beds well separated from shale. According to Holtedahl's (1912) descriptions of the nature and qualities of limestone in the Oslo Rift, a review largely based on Kiær's (1908) work on the sedimentary rocks of the region, the unit best fulfilling these criteria is the Silurian Borealis-Pentamerus Limestone in Hadeland. The second formation selected for sampling was the Ordovician Gastropod Limestone, which is also one of the purer limestones and the most easily available unit in the Hadeland area. Geology and sample locations are shown in Fig. 1.

The Palaeozoic sedimentary rocks are, in many places, intruded by Permian plutons. Samples were collected at variable distances from intrusive contacts in order to compare limestones with different degrees of metamorphic influence. Emphasis was also placed on following the strike and sampling the same level within a formation, for the purposes of comparing material which was as similar as possible prior to metamorphism. In several places, however, it was problematical to strictly follow the same horizon over larger distances. The samples, nevertheless, crudely originate from the same

level and should represent fairly similar original material. Most samples were collected in quarries and road-cuts with well-exposed stratigraphy.

Sample distances from the intrusions can be seen in Fig. 1. Around the small, northern gabbro intrusion, metamorphic effects were observed up to about 250 m from the contact, but distances vary. The contact aureoles around the batholiths in the south of the area can be up to 3 km (Dons et al. 1996, Svensen & Jamtveit 1998). Within an aureole, a similar degree of metamorphic alteration was not always to be found at quite the same distance, at surface level, from the intrusive contact. It was most adequate to label the samples based on a combination of distance and visible alteration effects and to group them into 3 broader categories; non-metamorphosed, low-medium metamorphosed and strongly metamorphosed limestone.

The Borealis-Pentamerus Limestone is of Lower Silurian age and comprises level 7a and 7b $\alpha$  according to the conventional stratigraphical nomenclature of Kiær (1908), Holtedahl (1912) and others. In a formal lithostratigraphic scheme it belongs to the Rytteråker Formation (Worsley et al. 1983). The Borealis Limestone of level 7a occurs as a massive bed of 4-5 m thickness. It is a bluish-grey biosparite characterised by the abundance of large, thick, white brachiopod shells of *Pentamerus Borealis* (Holtedahl 1912). Other fossils of importance are corals and stromatolites (Owen 1978). It is succeeded by

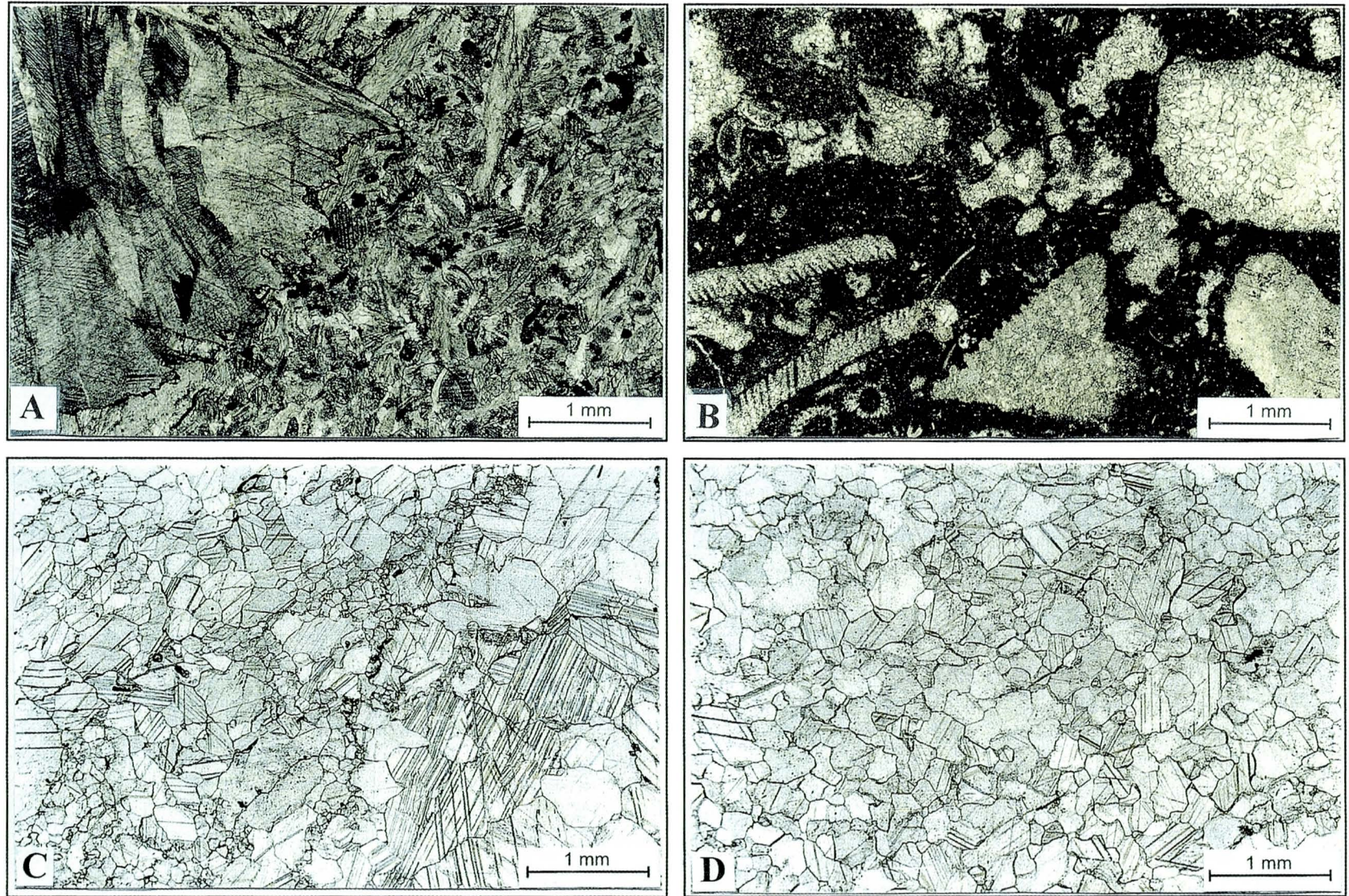


Fig. 2. Textures in the limestones. Contamination by carbon dust is prominent in the non-metamorphosed limestone, in contrast to the white and pure calcite in the contact-metamorphic limestone. The originally dissimilar limestones have comparable, recrystallised, crystalloblastic textures after metamorphism. (A) Non-metamorphosed Borealis Limestone: groundmass of sparry calcite, and large characteristic brachiopod shell in left part of photo. Sample P25, PPL. (B) Non-metamorphosed Gastropod Limestone: fossils set in a matrix of very fine-grained, dark brown micrite. Sample H4, PPL. (C) Strongly metamorphosed Borealis Limestone, from close to the intrusive contact. Sample P7, PPL. (D) Strongly metamorphosed Gastropod Limestone, close to the contact. Sample H30, PPL.

the related *Pentamerus* Limestone, level 7b $\alpha$ , which is a 5-7 m-thick zone of massive bioclastic limestone (Holtedahl 1912, Owen 1978). The *Pentamerus* Limestone differs from level 7a in lacking the thick shells; instead, some horizons contain abundant thinner and usually smaller pentamerids.

The Gastropod Limestone of Upper Ordovician age comprises level 5a and is named the Kalvsjø Formation in the new lithostratigraphical terminology (Owen 1978). It consists of nodular and bedded bioclastic limestone alternating with thin shale and siltstone layers; limestone forming 50-70 % of the formation (Owen et al. 1990, Braithwaite et al. 1995). In the sampled areas, the Gastropod Limestone is composed of typically 10-20 cm-thick nodular limestone beds interlayered with 1-3 cm-thick shale layers. The colour of the unaltered limestone is dark grey. It is richly fossiliferous, particularly in echinoderm and gastropod debris. Fragments of calcareous algae, brachiopods, corals, bryozoans and orthocones are also common (Owen 1978, Braithwaite et al. 1995).

The nature of the groundmass is different in the two limestone formations. The matrix of the non-metamorphosed Gastropod Limestone is entirely composed of very fine-grained, opaque or semi-opaque micrite (microcrystalline calcite) with a dark brown colour (Fig. 2B). In contrast, the non-metamorphosed level 7a *Borealis* Limestone has a relatively coarse-grained sparry calcite matrix (Fig. 2A). The groundmass in level 7b $\alpha$  is a mixture of coarse spar and very fine-grained, dark brown micrite, usually with the latter dominating.

Contaminating minerals in the limestones and their metamorphosed equivalents include quartz, pyrrhotite, pyrite, magnetite and calc-silicates like epidote, amphibole, pyroxene, garnet, wollastonite, vesuvianite, and titanite.

Svensen & Jamtveit (1998) have made estimates of the metamorphic conditions for the southeastern part of the area based on phase petrology. The peak temperatures of metamorphism associated with intrusion of the batholiths in the Grua area have been calculated to ca. 560°C, assuming a pressure of 1 kbar, and at distances 2 km from the intrusive contact a temperature decrease of ca. 170°C is suggested.

## Results

The main effects that contact metamorphism has had on the quality of the *Borealis*-*Pentamerus* and Gastropod limestones with respect to their textural properties, brightness and geochemistry, are presented below. More detailed descriptions of the petrographic observations, analytical data and results are given by Kjølle (1998).

Field relations and petrographic investigations have shown that an increasing degree of contact metamorphism generally is accompanied by a gradual change in the following textural parameters:

- The grain size increases
- The rock becomes lighter in colour
- A purification of the calcite with regard to the content of carbon dust
- Grain shape and grain boundaries become more even

Within the contact aureoles, the character of the limestones varies from relatively fine-grained and dark grey in colour near the outskirts of the aureoles to medium-grained or coarsely crystalline marble with medium to light grey or almost white colour approaching the intrusions. Figure 2 shows some of the textural features. Thermal metamorphism has resulted in a penetrative recrystallisation of the limestone to a crystalloblastic texture. The smaller grains in particular, but also the coarser grains, form a polygonal mosaic with regular grain shape and even grain boundaries. Most common grain sizes in the most contact-affected limestone are 0.2-2 mm. The greater part of the calcite in the non-metamorphosed limestone has a brownish colour and appears impure due to a dusting of cryptocrystalline inclusions. These dark particles are most probably carbon dust. The dark grey colour of carbonate rocks is generally attributed to organic compounds, especially very finely dispersed carbon dust (Vogt 1897, Dons 1977, Blatt 1982, Todd 1990). The content of such dust particles is much reduced in the contact-metamorphosed limestone. The calcite grains become clearer, lighter and purer with increasing intensity of contact metamorphism.

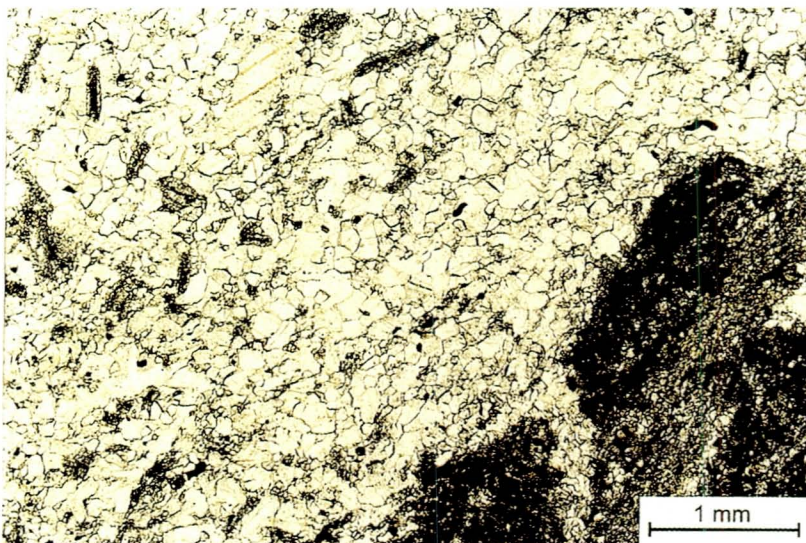


Fig. 3. Occurrence of the impurities after contact metamorphism, shown by a sample of medium metamorphic Gastropod Limestone. Contaminants are mainly calc-silicate minerals. These are, in part, present as scattered small crystals, which can occur along grain boundaries but also intergrown with the calcite; and, in part, they are present as very fine-grained, turbid masses that invade and replace the calcite. With regard to carbon dust content, the calcite in this photo is relatively light and pure compared with much of the medium-metamorphic calcite. Sample H22, PPL.

Exceptions to these textural changes do, however, occur. Contact metamorphism usually leads to a grain size increase in biomicritic limestones, but this is not necessarily the case for carbonates with spar matrix. The non-metamorphosed Borealis Limestone is of similar coarse grain-size as the strongly metamorphosed equivalent. There are also examples of biomicritic limestone which has neither increased in grain size nor been bleached during the metamorphism, as shown by some low- to medium-metamorphosed samples of the Gastropod Limestone. This variety has an almost similar texture to the non-metamorphosed equivalent, although it is more recrystallised. At some fossil sites recrystallisation can be seen to have resulted in a finer rather than coarser grain size; cf. Fig. 6 in Kjølle (1998).

Of particular importance for the industrial mineral quality is the amount of impurities and how these occur in the rock. In these respects the two investigated limestone formations show no improvements in quality after contact metamorphism, with the exception of the reduction in abundance of carbon dust. The amount of all other observable contaminants is apparently unchanged or has increased. Metamorphic recrystallisation of the original constituents is the most common, with alteration of shale inclusions to calc-silicates. In some places, an increase in the amount of impurities can be seen, where the marble has undergone metasomatism (addition of new elements from an external fluid) and the calcite has been replaced by masses of calc-silicates (skarn) ± sulphides.

The occurrence of the contaminants appears to be directly comparable in the non-metamorphosed and contact-metamorphosed limestone. In unaltered as well as metamorphosed samples, impurities are present both as evenly spread grains and segregations, commonly along grain boundaries, but also intergrown with, or enveloped by the calcite. Even though contact metamorphism produces more even grain-shape and -boundaries and increased grain size for the calcite, the impurities can be just as fine-grained and commonly intervene in the calcite to a similar degree in metamorphosed as in non-metamorphosed limestone. Figure 3 illustrates the occurrence of the impurities in the marble. The calc-silicate contaminants are present both as scattered small crystals and as very fine-grained, turbid masses or 'veils' that invade and replace the calcite.

Geochemical analyses, including XRF, acid-soluble CaO and MgO, and total organic carbon (TOC), as well as measurements of brightness, were made on most of the samples (Kjølle 1998). With regard to chemical composition, the majority of detected elements show no systematic change with the degree of metamorphic influence. In the Borealis-Pentamerus Limestone nearly all elements occur in comparable amounts in the non-metamorphosed and

strongly metamorphosed samples. In the Gastropod Limestone however, a weak change is seen for Fe<sub>2</sub>O<sub>3</sub>, CaO, SiO<sub>2</sub>, K<sub>2</sub>O, Ba, Rb, Ni and V. An almost unmodified chemical composition is in accordance with the textural observations. (It should, however, be noted that in order to best evaluate purely metamorphic effects, sampling of limestone which appeared metasomatically altered was avoided). This result is also in agreement with the general view and with other studies of limestones indicating that metamorphism is essentially isochemical (insignificant change in chemistry) (e.g. Einaudi et al. 1981, Best 1982, Labotka et al. 1988).

The brightness values measured on ground samples are shown in Fig. 4. The sample powders represent a similar degree of grinding with much the same distribution of grain size (Kjølle 1998), indicating that their brightness values can be compared. Figure 4 clearly shows that contact metamorphism has increased the brightness of the limestones. The non-metamorphosed Borealis-Pentamerus Limestone has a reflectivity of 50-70 %. The lower values represent level 7bα,

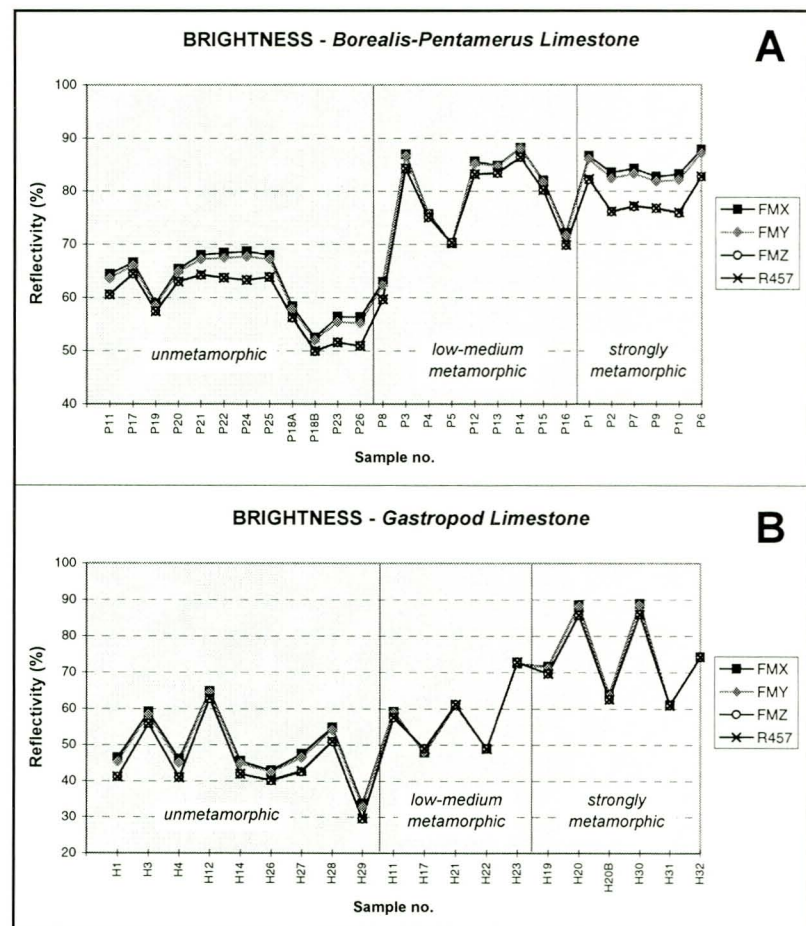


Fig. 4. Brightness of non-metamorphosed and contact-metamorphosed limestone, A. Borealis-Pentamerus Limestone, and B. Gastropod Limestone. The measurements of reflected light have been made with 4 filters. The brightness is given by R457, which is the reflectivity of monochromatic light of wavelength 457 nm. FMX, FMY and FMZ are the reflectivities of light of wavelengths centred around 600 nm (red), 550 nm (green) and 450 nm (blue), respectively.

in agreement with a slightly darker colour of these samples. The weak- to medium-metamorphosed samples range from 60 to 88 % and overlap with the strongly metamorphosed samples, which lie within the range 76-88 %. The Gastropod Limestone shows more of an increasing trend with the degree of metamorphism; brightness values being 30-65 % for non-metamorphosed, 48-73 % for weak-medium metamorphosed, and 61-89 % for strongly metamorphosed samples.

## Discussion

It is a common conception and often true that the grain size of a rock increases with metamorphic grade, but this effect is not universal and need not apply to all minerals present. Ascribing metamorphic grain size just to temperature-pressure conditions can be too simplistic and very misleading, because grade is only one factor influencing grain size (Vogt 1897, Best 1982). In marbles, impurities of calc-silicates can be fine-grained in some situations and coarse in others. Metamorphic calc-silicates are typically fine-grained, whereas calc-silicates formed metasomatically commonly occur as coarse crystals (Einaudi et al. 1981). Metamorphic recrystallisation of the Borealis-Pentamerus Limestone and Gastropod Limestone resulted in a grain size increase for the calcite, whereas the impurities remained fine-grained and partly intergrown with the calcite. Even metasomatic calc-silicates can occur with very small grain size (a few  $\mu\text{m}$ ), as for example those formed in contact-metamorphosed carbonates in the Ravndalskollen-Myrerollen area of the Oslo Rift (Jamtveit et al. 1992a).

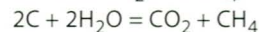
For a number of limestone uses, e.g. as filler, impurities first have to be liberated by grinding and removed by beneficiation processes. The occurrence of impurities in the limestones from the present study does not indicate that contact-metamorphosed limestone would be simpler to purify than unaltered limestone. Although some limestones may have experienced grain coarsening of the contaminating minerals as well as the calcite, and movement of most impurities to calcite grain boundaries, this study illustrates that one should not take for granted that contact metamorphism will automatically produce such quality improvement.

With regard to the colour of the limestone, there is clearly a connection with the measured brightness values and the amount of carbon dust observed in the calcite under the microscope. Metamorphism clearly bleaches the rock as well as increasing the reflectivity values of the ground samples. There is an excellent agreement between the relative degrees of bleaching of the hand specimens and the relative brightness values. Furthermore, when samples of roughly similar grain size are compared, it can be seen that the darker the rock colour, the more contaminated are the calcite grains by dark carbon dust particles. The lowest content of carbon dust in the calcite is observed in the most strongly contact-affected samples with the lightest colour. These relations imply that limestone with the strongest metamorphic imprint most probably has the lowest content of organic carbon. The exact amount of organic carbon is unknown; it

could not be measured using the TOC method. On the other hand, the detection level of 0.10 wt. % indicates that the amount of organic carbon needed to give limestone a dark colour is very small.

The colour of limestones partly depends on the grain size. They commonly have a darker colour the more fine-grained they are, in part because organic matter can be more finely dispersed in rocks of small grain size. In addition, micrite, which is analogous in hydraulic behaviour to clay minerals, adsorbs, and usually contains greater amounts of organic matter than do coarser grained limestones (Blatt et al. 1980, Blatt 1982). The dark colours of carbonate rocks are mainly attributed to organic matter, but sulphides and silicates can also contribute to the rock colour to some extent, as shown e.g. by sulphide-poor samples being slightly lighter in colour than samples containing more sulphides within samples of the same metamorphic grade.

Todd (1990) has studied bleaching of contact-metamorphosed limestone at Notch Peak, Utah, and relates the colour alteration in the limestone to organic carbon. His TOC analyses show that bleached samples contain less organic C than dark samples and point to loss of organic C during metamorphism. Carbon is interpreted to have been removed by a water-rich fluid. Phase relations for C-O-H fluids under metamorphic conditions set a limit to the mole %  $\text{H}_2\text{O}$  that can be in equilibrium with graphite. If  $X_{\text{H}_2\text{O}}$  is increased beyond this limit, the fluid will react with the graphite and carbon is removed as  $\text{CO}_2$  and  $\text{CH}_4$  according to the reaction:



Influx of a  $\text{H}_2\text{O}$ -rich fluid during metamorphism thus seems to be a possible mechanism by which limestone can be cleansed of organic carbon. This is supported by the data of Labotka et al. (1988) which indicate that argillaceous layers within the same formation were infiltrated by substantial volumes of  $\text{H}_2\text{O}$ -dominant fluids from the Notch Peak intrusion during the contact metamorphism.

Despite substantial fluid infiltration during contact metamorphism at Notch Peak, the limestone-shale sediments were apparently not metasomatised; their primary composition being essentially preserved (Labotka et al. 1988). Contact metamorphism of limestone-shale sedimentary rocks in the Oslo Rift was also to a large extent driven by through-flow of  $\text{H}_2\text{O}$ -rich fluids (Jamtveit et al. 1992b). It is common, however, both in the Oslo Rift and elsewhere, that contact metamorphism is locally accompanied by metasomatic skarn formation. In the study area, good exposures of limestone can be seen which are extensively replaced and contaminated by calc-silicates and sulphides, for example in road-cuts and quarries at Grua. Therefore, one should be aware that certain zones of contact-metamorphic environments are likely to have had addition of new components from an external source and, consequently, contain limestone which is less pure than in its primary state.

Finally, it has been noted that also the mechanical properties of the limestone seem to vary with the degree of contact metamorphism. The non-metamorphosed and the weak- to medium-metamorphosed limestone appear more solid and fresh than the most contact-affected analogue. The

latter seems more sugary, crumbles more easily, and is more readily weathered. This is possibly connected with the polygonal foam texture; even grain boundaries are best developed in the most metamorphosed limestone. It appears, therefore, that contact metamorphism has a disadvantageous effect if the marble is intended for natural stone applications. This is in full agreement with Vogt's (1897) conclusions, based on studies of a range of Norwegian marbles.

## Conclusions

The investigations have shown that contact metamorphism can, in some respects, have a favourable effect on the industrial mineral quality of limestone. It can reduce the amount of organic matter (carbon dust), giving the rock a lighter colour and increased brightness values, and it can produce more even grain-shape and -boundaries and increased grain size for the calcite. However, there are no indications from the present study that contact metamorphism improves the quality in terms of the occurrence and abundance of contaminants. These limestones show that the impurities can remain very fine-grained and partly intergrown with the calcite, implying that contact-metamorphosed limestone may be no simpler to purify than non-metamorphosed limestone. If the marble has also been metasomatically altered, which is not uncommon in contact-metamorphic environments, the rock is likely to be more contaminated and therefore of poorer quality than it was prior to metamorphism. Overall, the results indicate that contact metamorphism may not improve the limestone quality sufficiently to produce an economic industrial mineral deposit. It may be required that the limestone was of very high purity prior to metamorphism and also has escaped any accompanying metasomatism.

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