The Sodalite Group Minerals

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CHAPTER 1

Introduction

The start for this study and the resulting eBook was an unusual luminescence in a piece of photochromic sodalite (HPE specimen) from the Koksha area in Afghanistan. You can see the effect in this short Youtube movie. Because more minerals in the sodalite group have special effects, the study has been expanded to the “common” minerals of this family. Unless otherwise mentioned all photographs and images in this book are made by the authors.

A first acknowledgement has to be directed to all Belgian and Dutch collectors that gave specimens for photography or even destructive research:

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Both authors are member of the Mineralogische Kring Antwerpen (MKA). Details to be found on their website.

CHAPTER 2

Group Overview

The “common” (and recognized) minerals we were referring to in the introduction are:

sodalite	$Na_8\ (Al_6Si_6O_{24})\ Cl_2$
haüyne	$Na_6Ca_2\ (Si_6Al_6O_{24})\ (SO_4)_2$
nosean	$Na_8\ (Al_6Si_6O_{24})\ (SO_4).H_2O$
lazurite	$(Na,Ca)_8\ (Al_6Si_6O_{24})\ S_2$

The table only shows the ideal end members. The literature often refers to more complex compositions, but that is a story for later. Haüyne and nosean form a complete solid solution series and also the other end members can mix. Lazurite is also a seperate story. Its possible existance is questioned by certain groups. We will give our vision in this book.

Other members are hackmanite, a **photochromic** sodalite, and lapis lazuli, a blue lazurite variety used as a gemstone. Both are no longer official mineral names (see IMA mineral list) and lapis lazuli is more a rock than a mineral. We also use the spelling of the mineral names used in that list.

The sodium in the formula can be partially replaced by potassium or calcium. They are used in an alternative system to distinguish the members.

We will also show that the typical orange **luminescence** under UV-A can be used to distinguish between the members. We will show typical examples.

The above table lists the “common” minerals of the sodalite group. The others (more rare) are made through typical ion-exchange. E.g.

$Al^{3+} + Na^{+} \rightarrow Be^{2+} + Mn^{2+}$: Helvine series

$2\,Al^{3+} \rightarrow Be^{2+} + Si^{4+}$: Tugtupite series

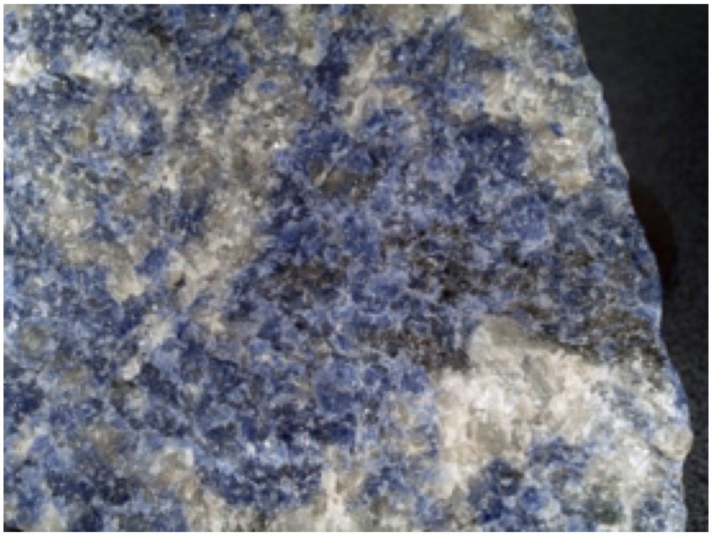
Details about the **spectrometers** used can be found in chapter 8. We use the scientific terminology for luminescence. That is partly different from the generally used terms by mineral collectors. See chapter 9 for details.

CHAPTER 3

Applications

Besides the gemstone applications of the very blue varieties, the intense blue minerals (especially lazurite) were used to prepare the natural dye **ultramarine**. It was very expensive because the purification process out of lapis lazuli was extremely time consuming. Nowadays ultramarine is made synthetically. Many articles can be found rearding the synthesis of lazurite out of cheap chemicals. It used to be called the “most perfect colour”.

There are also expensive floor and wall tiles for sale made of sodalite (Ernst Burke, private communication) under the trade name “Azul Bahia”. It concerns a sodalite quarry in the Bahia province in Brazil where this rock (sodalite **syenite**) was mined (Lourdes 2005, 2007). A typical price I found on the internet was around 75 EUR/m2.

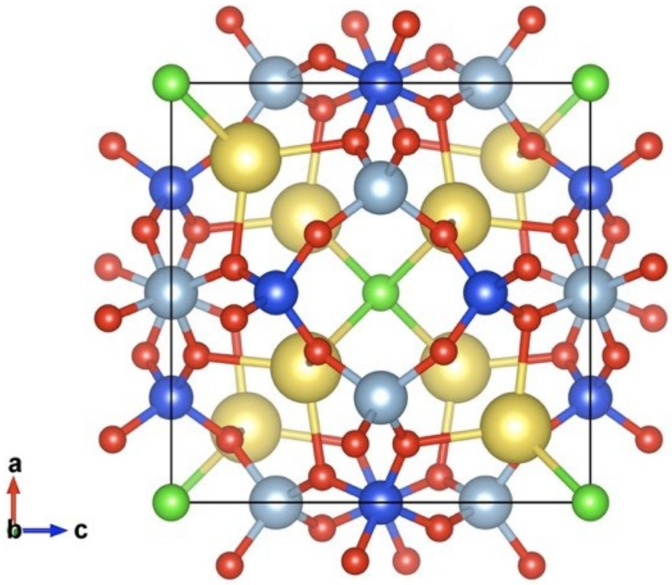


Sodalite syenite: Sodalite and alkali feldspar together with biotite, Floresta Azul. An alkaline complex, Bahia, Brazil (coll. EBU)

Another possible application was based on the photochromic effect. Some varieties can be coloured by UV light and be bleached by visible light (more details in chapter 6). Because of possible applications for display screens, many studies are published on synthetic sodalite. Up til now, there is no practical use.

CHAPTER 4

Crystal Structure

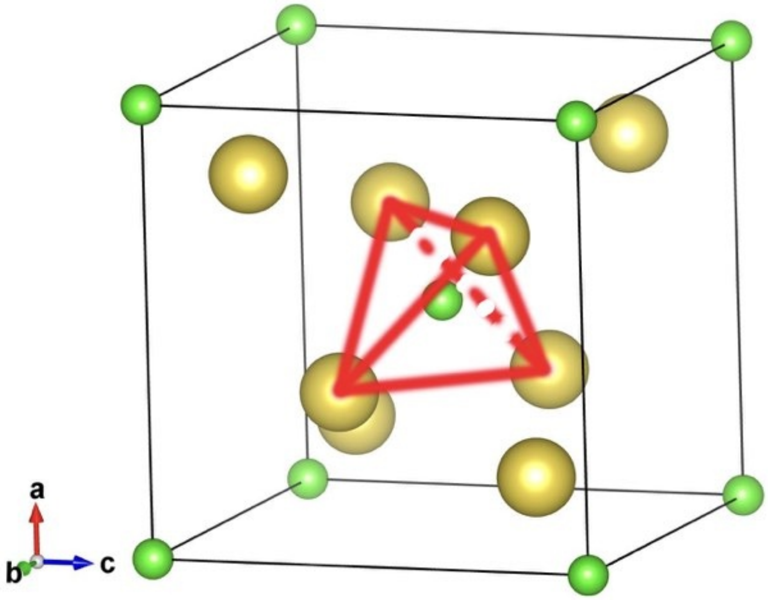


Crystal structure of the sodalite group. Blue: Silicon, Silver: Aluminium, Yellow: Sodium, potassium or calcium, Green: Chlorine, sulphate, sulfide, water , …, Red: Oxigen. Drawing made with VESTA (Momma, Izumi, 2011)

Because it is a **silicate**, the basic building block will be the SiO_4 tetrahedron. They are linked to form a **zeolite** like network. Half of the silicon (valence 4) is replaced by aluminium (valence 3) to compensate the cation charges (Na, Ca, K). The tetrahedrons containing Si and Al are ordered.

The cages.

Important for the further chapters is that sodium (or its replacing cations) forms tetrahedral cages in which chlorine (or the replacing groups) is captured. This yields a [Na^+_4 Cl^-] complex or equivalent with other ions.



The sodium cage (red lines) in which the chlorine (green) is captured. The silicate lattice is not shown. Drawing made with VESTA (Momma, Izumi, 2011)

The lattice.

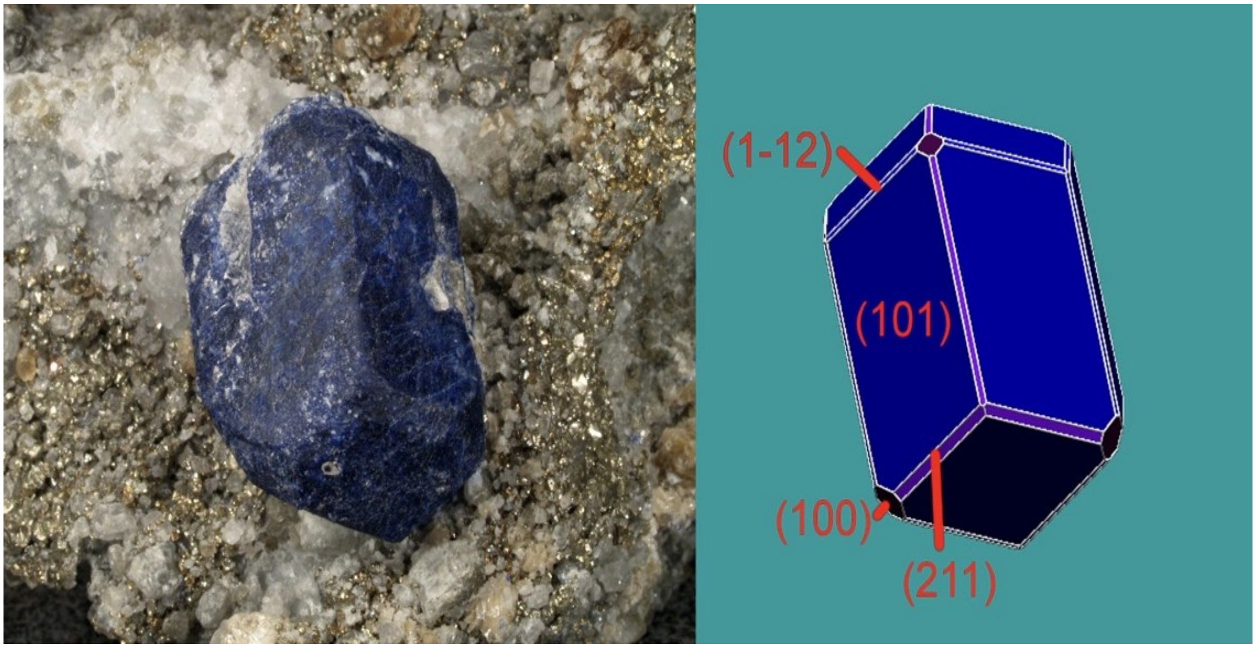
Sodalite type minerals are cubic having **point group** -4 3 m and **space group** P-4 3 n. The **lattice parameter** is variable and dependant on the ions (chlorine is smaller that the sulphate group).

But there exist orthorhombic and even lower symmetries (see e.g. Tauson, 2012 for Lazurite) because of ordering of the sulphur groups or other present ion-groups (Tauson, 2014, Hassan, 1989).

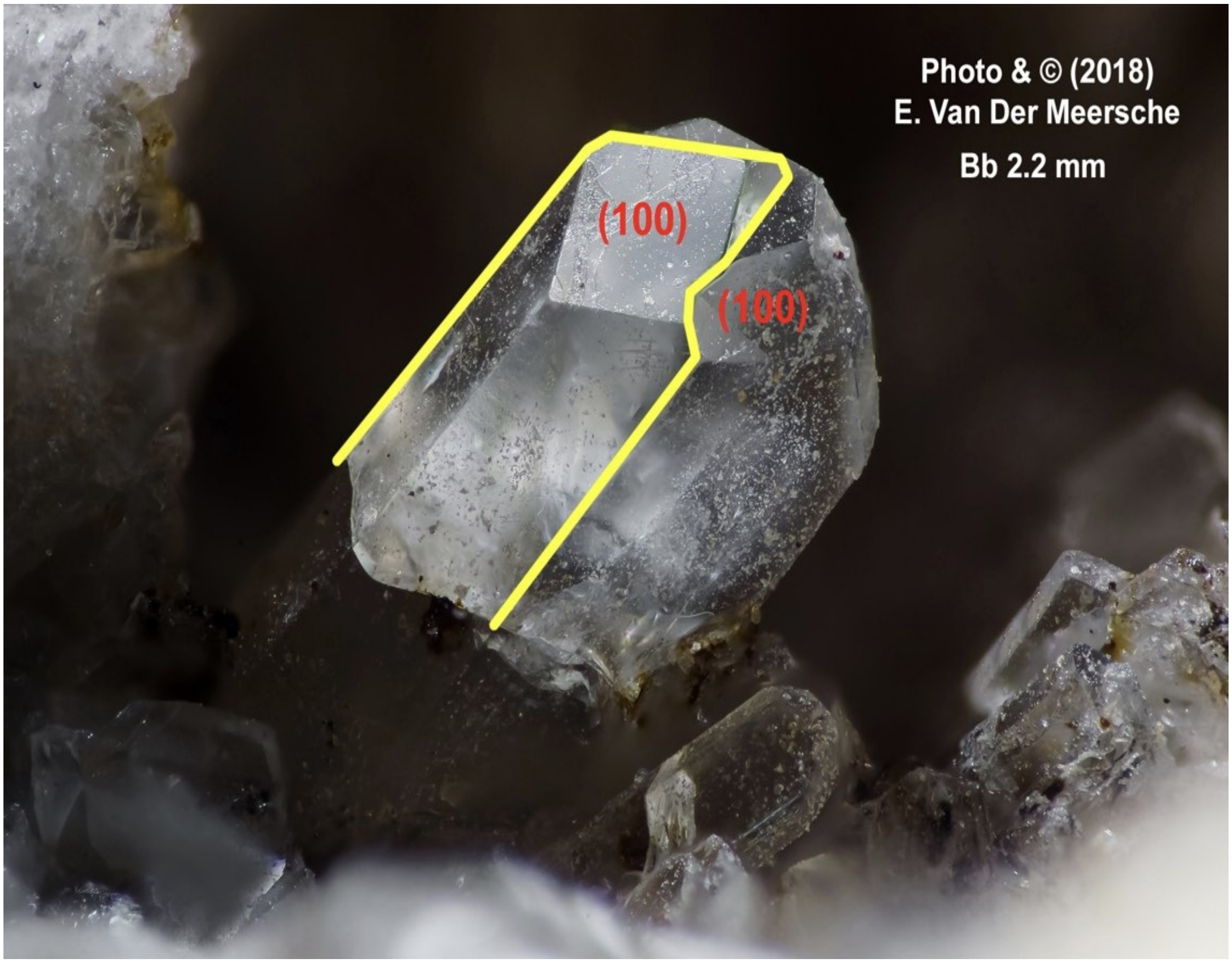
	Lattice parameter (nm)
Sodalite	0.888 (0.887-0.888)
Nosean	0.907 (0.905-0.908)
Lazurite	0.907 (0.907-0.909)
Haüyne	0.911 (0.908-0.913)

Crystal habit.

Crystals usually have a **dodecahedral** shape, often elongated in a [111] direction. **Twinning** occurs on one of the (111) planes (octahedral), mostly parallel twins. They can be recognized by the fact that there are 6 dodecahedral planes at the long ends of the crystal and not three and very often you see concave corners. The dodecahedral planes that form the prism coincide in both individuals.



Lazurite crystal from Sar-e-Sang, Koksha valley, Afghanistan (RDE). Note the presence of pyrite in this typical paragenesis.



Nosean twin from Laacher Kessel, Mendig, Eifel, Germany. The individuals are seperated by a yellow line. Note the 2 small cube planes (100) in each indivudual 60° apart. Also note the concave angle at the upper yellow line between the cube faces. (EVDM, also picture)

CHAPTER 5

Chemistry And Occurrence

Sodalite.

Pure sodalite is the chlorine end member of the group. Pure sodalite is white, but can get almost any colour. As with most silicates, the strike is white. Hardness is between 5.5 to 6.

Sodalite analysis found in the literature and in the RUFF database (Lafuente , 2015 and http://rruff.info/), are all very close to the pure end member. Sometimes the sodium is replaced by vacancies or potassium, but always only some 1 to 2 %.

Sodalite is a typical mineral in alkali rich **plutonic rocks** and associated **pegmatite** (e.g. nepheline sodalite, Andersen, 2013). It is also found in **metamorphic** limestone rich rocks that were in contact with alkali rich volcanic rocks.

A seperate word about “hackmanite”, a sodalite variety where part of the chlorine is replaced by sulphur (in sulfide form). Hackmanite is photochromic and exhibits a typical luminescence. Both effect will be treated in detail in later chapters. Hackmanite is no longer an official mineral name (IMA master list)

When is a sodalite a “hackmanite” ? Many definitions exist (see also Mindat)

- Sodalite containing sulphur and exhibiting an orange luminescence

Sodalite having photochromic properties.

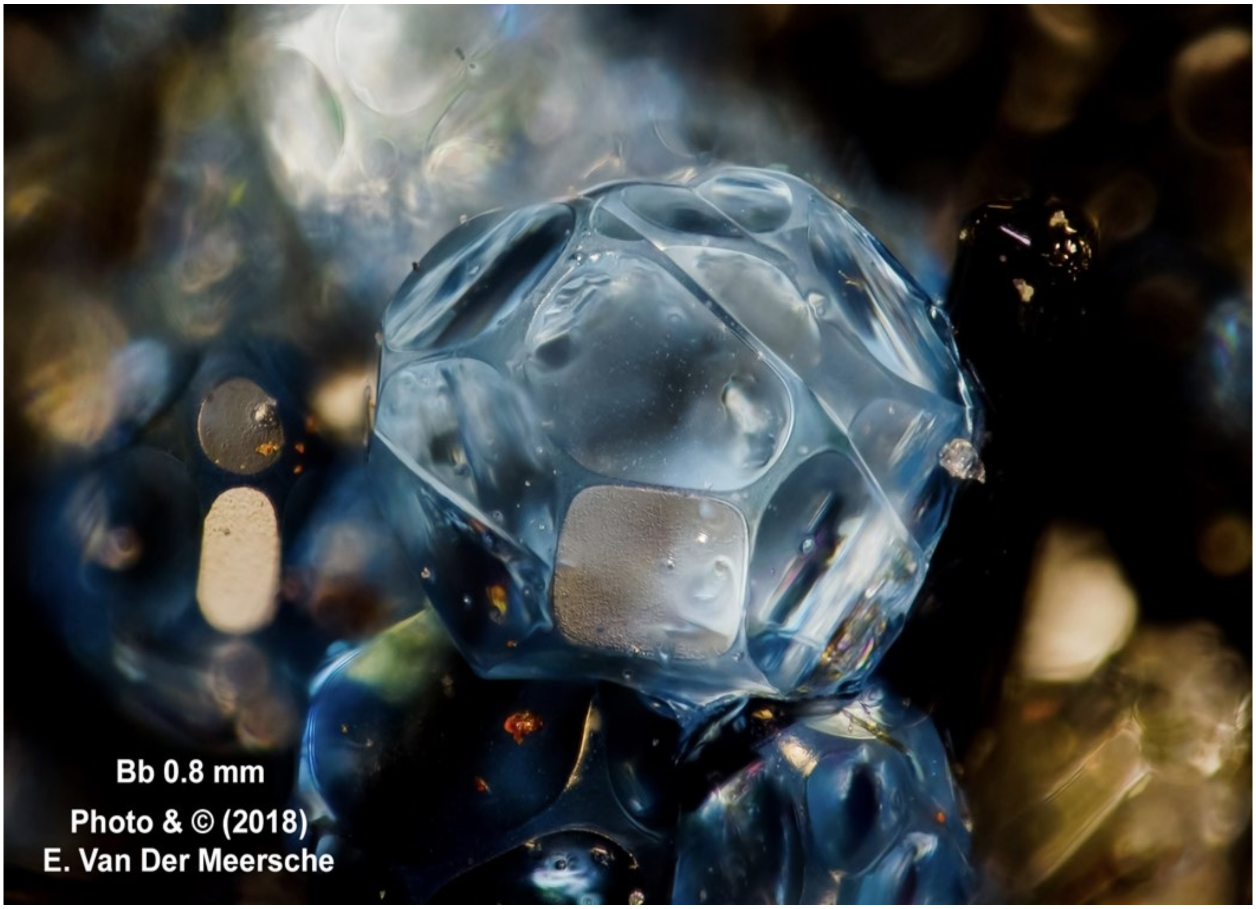
- A white to grey sodalite with traces of sulphur (<1%)
- A combination of the above properties

Personally we favour the photochromic definition. This is also found in recent literature (Norrbo , 1916 and Finch , 2016). But instead of using “hackmanite”, a better description, conform IMA, would be photochromic sodalite.

Haüyne and nosean.

Haüyne and nosean are the sulphate end members of the group. Haüyne is the pure sulphate end member. In ideal nosean half of the sulphate is replaced by water. A complete solid solution exists. Solid solution towards lazurite (next paragraph) is also possible.

A nosean listed in the RUFF database contained besides sodium also calcium, vacancies and potassium. Sulphate/water ratio was 50/50.



Haüyn, Krufter Ofen, Kruft, Eifel, Germany (EVDM, also photo)

Blue nosean in sanidinite, Laacher Kessel, Mendig, Eifel, Germany (EVDM, also photo)

Good examples of solid solution can be found in the literature. Rocks from Mt. Vulture in Italy contain different members of the group (Di Muro, 2004). A black haüyne contains besides sulphur also 25% chlorine (*). White nosean is described containing 50% sulphate and 25% chlorine. Both clearly in the sodalite direction. The Vesuvius complex contains sodalite, haüyne and nosean (Ballasone, 2016). Sodalite appraches the pure composition, but the chlorine contant of nosean (15 to 42%) and haüyne (15 to 30%) is still high and also haüyne contains water.

Nosean is typical for undersaturated **phonolitic** rocks. It is rarely found in non-volcanoc rocks, except in aegirine-augite syenites. Haüyne is also typical for phonolites.

(*): The percentages here and in the following paragraphs are expressed towards the 2 anions in the formula. They count for 100 % (e.g. If there are 2 chlorines present, then chlorine is 100% present)

Haüyne is blue and nosean is white ?

That has longtime been stated and used as a differentiation. A detailed study by Cruse and Hentschel, 1990, proved this to be wrong. We have the same findings when using the sulfide vibronic emission spectrum (see chapter 6). White or clear haüyne is possible, as is blue nosean (photo previous page).

Transparent, clear haüyne crystal (twinned) from Laacher Kessel, Mendig, Eifel, Germany (EVDM, also photo)

Intermezzo: Haüy In Paris

René Just Haüy (1743-1822) was a famous mineralogist that contributed a lot to geometrical crystallography. He became a priest but in his free time he was interested in botanics and mineralogy.

After a scientific career in many institutions, he became lecturer at the museum d’histoire naturel in Paris (1802-1822). The mineralogy department of this museum possesses a big statue of him in white marble. It is located in the Jardin des Plantes in Paris. The mineral haüyne was named after him.

The museum is worth a visit since it renewed the exposition of minerals in an interactive exhibition. Many topics of mineralogy and gemmology are shown in different windows. The museum also has some nice pieces of the sodalite family.

Other impressive topic are extremely large crystals (of several 100 kg) of different minerals.

Left: the impressive archive of mineral specimens. Right: Massive mineral crystals dominate the central part of the exhibition.

We were able to get access to their archive collection to make a study of the different samples of the sodalite family in their possession (cfr. acknowledgements).

For more info on the museum, its departments and the opening hours, consult their webpage.

Lazurite: lazu-right or lazu-wrong ?

Lazurite is story apart. The pure sulfide end member has not been found yet in nature. It is also not easy to make synthetically. Most of it still contains a high percentage of sulphate (Fleet, 2005). Wagner (2012), reports a synthetic sulfo-sodalite, only containing sulphur (in different configurations).

As a result, there is discusion in the mineralogical world about the name lazurite. Certain claim that natural lazurite actually is nothing more than sulphur containing haüyne or nosean. For future discussions, it is important that you remember that the sulphate and sulfide group are in the cages formed by the cations.

Fleet, 2005, uses the following definitions:

Lazurite if sulphate + sulphur = 1 or 2 atoms in the formula

- Haüyne if sulphate <= 2 atoms in the formula and sulphur almost zero

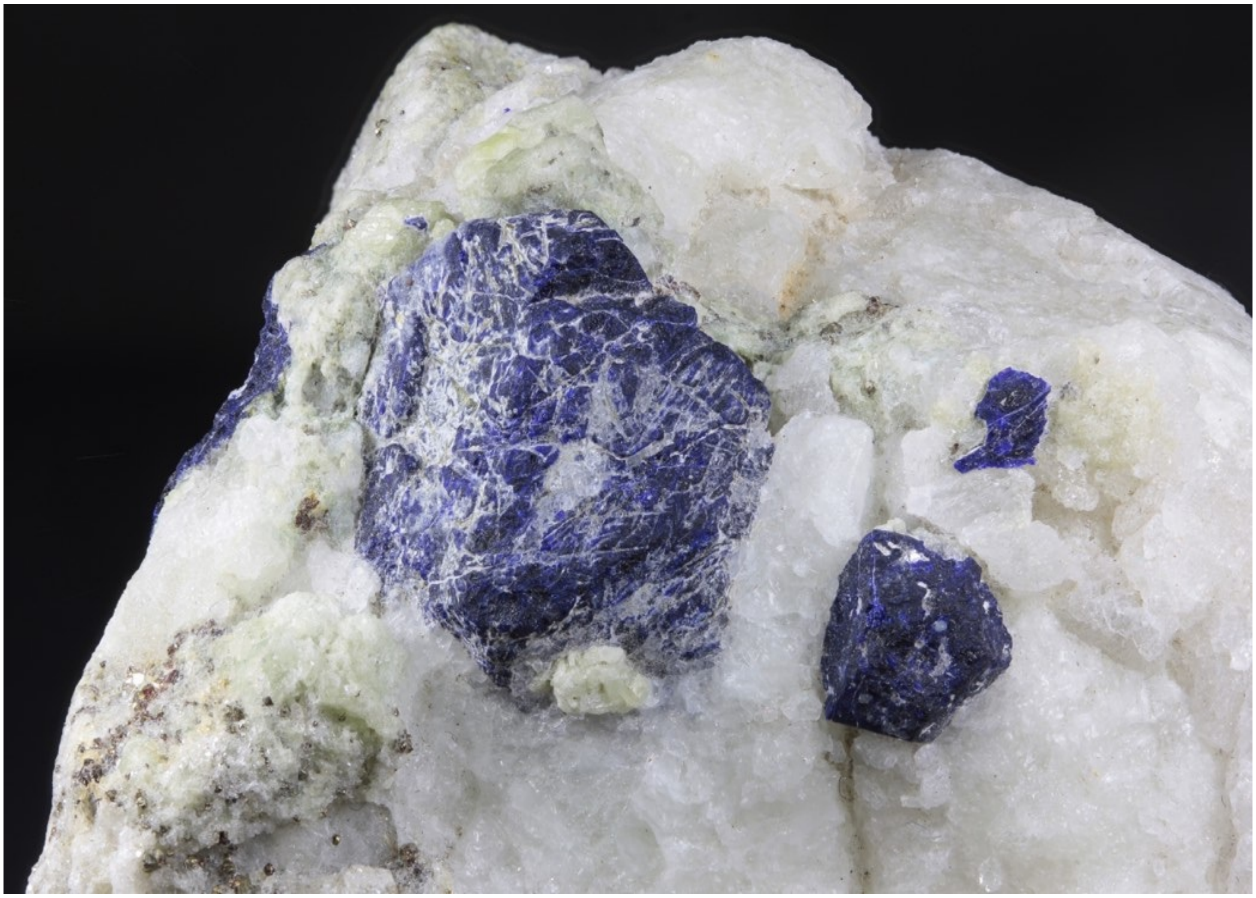
Sodalite if sulphate + sulphur < 1 atom in the formula, even without chlorine.

A lazurite in the RUFF database contains 67% sulphate, 32% sulphur and 1% chlorine. The 2 lazurites in the IMA reference (Hassan, 1985) contain resp. 22% (Afghanistan) and 43% (Baffin Island) sulphur. Other literature values from different localities (Afghanistan, Baffin Island, Baikal, Mt Vulture) range from 8 to 43% (Fleet, 2005, Tauson, 2011, Di Muro, 2004).

The sulphate/sulphur ration is influenced by the chemical environment. Reducing conditions favour sulphur, oxidising conditions favour sulphate. Furthermore, below appr. 550°C the cages become less mobile and the large groups (like sulphide and sulphate) are locked up. As such the ratio gives more details about the formation conditions (Tauson 2011, Di Muro 2004)

Raman spectroscopy is also used to distinguish between haüyne and lazurite (Wulff-Pedersen, 2000 in La Palma material). They use the terminology haüyne$_{SS}$ (solid solution) for lazurite to distinguish both minerals.

Lapis Lazuli refers to an intense blue variety of lazurite. It is often accompanied by pyrite and a white mineral that can be a silicate (very often wollastonite) or calcite. Typical occurrences are metamorphic limestones or contact zones between limestone and syenitic pegmatites.

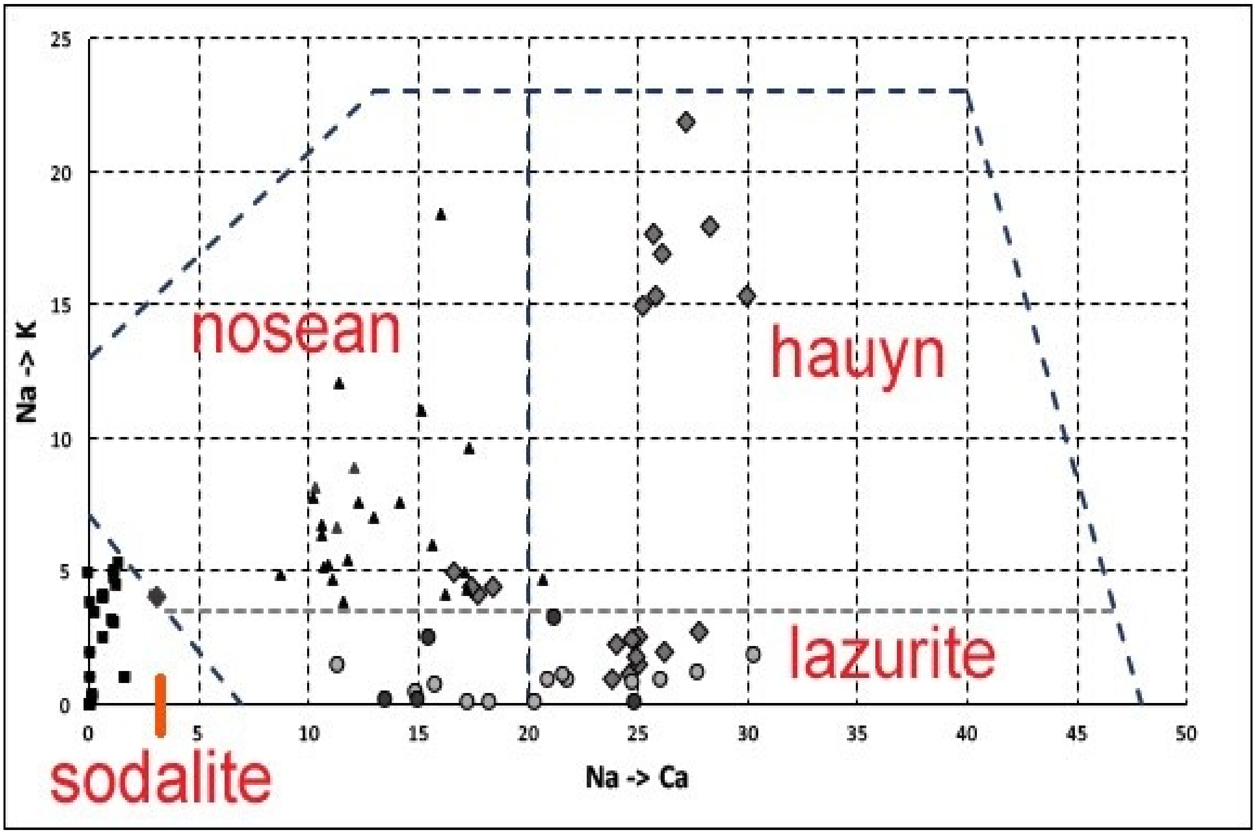


Lazurite crystal from Koksha, Afghanistan (LVG)

The Na- Ca- K triangle.

Some groups (Ballasone 2016, Di Muro 2004, Wulff-Pedersen 2000) use the Na-Ca-K ratio to distinguish between the minerals and it seems to give a better relation than the anion division, although exceptions also exist. Using this method is not that abnormal, since it is also used in other silicate groups (feldspars, zeolites, …)

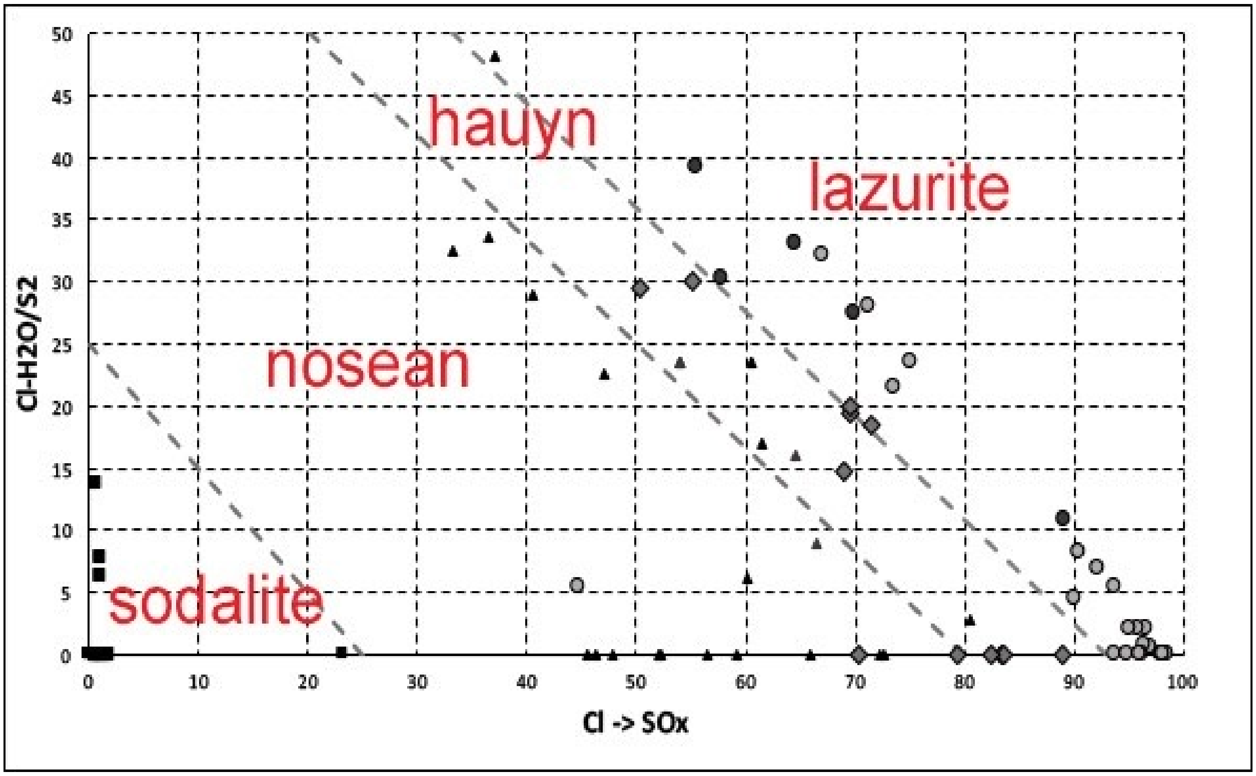
All analysis found in the literature (Hogarth 1980, Tauson 2011, Ballasone 2016, Di Muro 2004, Wulff-Pedersen 2000) are put in Na-Ca-K diagram. The limits used are those from Lessing (1971), except the lazurite line which was defined by using the terminology in the articles.



Na-Ca-K diagram. The x-axis represents increasing Ca-content, the y-axis potassium. The origin is 100% sodium. Squares: sodalite, triangles: nosean, diamonds: haüyne, circles: lazurite (dark ones: synthetic)

Sodalite lies in a clearly defined region. The other minerals too, but exceptions can be seen: haüyne from Mt. Vulture in the nosean area and haüyne from La Palma in the lazurite area (but that was the origin of the article). Lazurite is typical for a low potassium area.

A same diagram can be constructed using the anions. One axis is the oxidising axis (sulphate) and the other axis represent the other groups and reducing elements (sulphur).



Sulphate vs. other groups. Squares: sodalite, triangles: nosean, diamonds: haüyne, circles: lazurite (dark ones: synthetic)

Here there is more confusion. And as expected, the borderline between haüyne and lazurite is not very well defined. Furthermore, you need special analysis techniques to distinguish between the different oxidation states of sulphur. That explains the many points lying on the x-axis.

A division based on the Na-Ca-K ratio instead of the historically based anion groups is certainly worth a discussion within the IMA. Deviations can be given a prefix as is done in other silicates (e.g. pyroxenes, amphiboles) or is already done in the case of hydroxy-sodalite and carbonate-sodalite.

Lazurite is then potassium poor, haüyne potassium rich. Nosean is calcium poor, haüyne calcium rich. Lazurite containing a lot of sulphate can be named sulphate-lazurite or sulfide-haüyne. The name lazurite can be maintained in case a very sulfide rich variant is found.

We only have to wait until the IMA decides to initiate a workgroup around this theme. Discussion guaranteed.

CHAPTER 6

Colour Centres

Regarding colour in the sodalite group, a lot has been published. On certain points, authors agree, but on others there is still discussion. In this chapter, an overview.

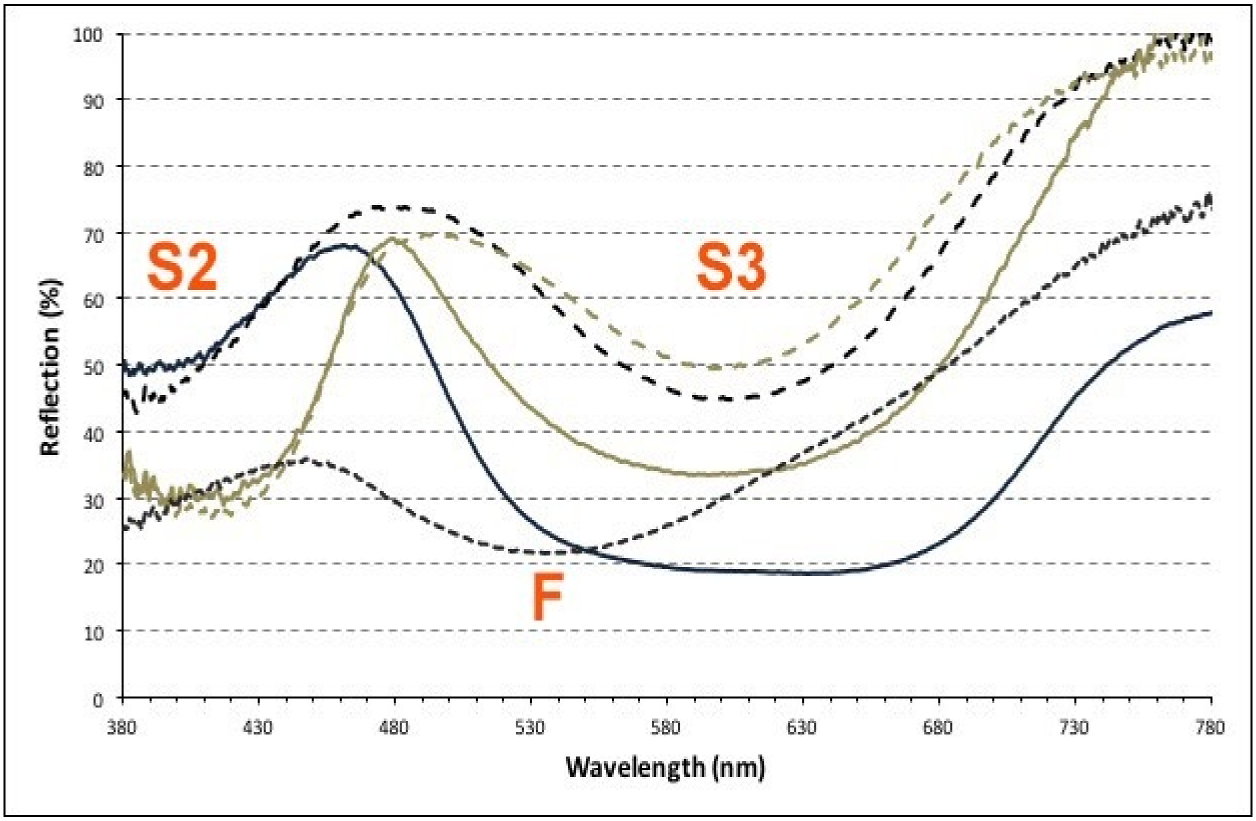
A general introduction to colour centres can be found in:

https://www.britannica.com/science/color-center

Sulphur.

Sulphur absorption.

Sulphur can be present in a number of states. S_2 (sulfide) and S_3 (tri-sulphur) cause a blue colour. Replacement of sulphur by selenium in synthetic sodalites gives a red colour (Schlaich, 2000). Finch (2016), attributes a yellow colouration to S_2 (absorption around 400 nm), S_3 for blue (absorption centered around 620-650 nm) and S_4 to purple (absorption around 510-520 nm). Norrbo (2016), also refers to S_3 at 600 nm. Two groups (Fleet, 2005 and Tauson, 2012) state that the link between blue and sulphur is not unambiguous yet.

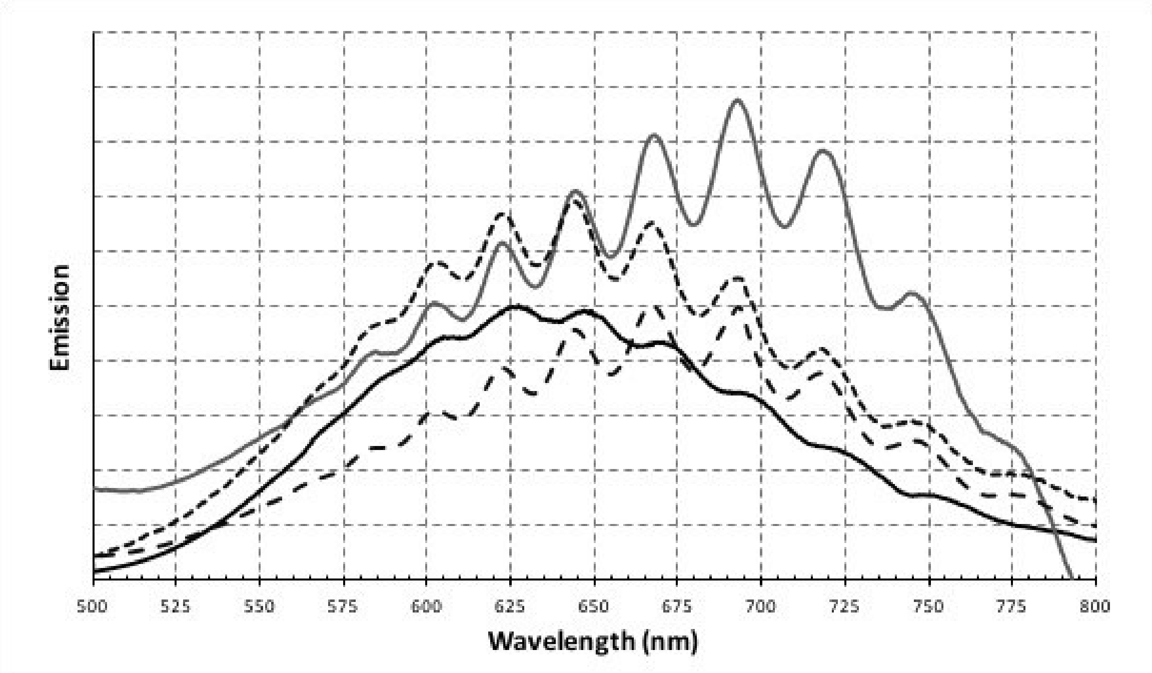


Reflection spectra of sodalite group minerals. All have the S3 absorption. Greenish colours have a stronger S2 absorption. The F-center, responsable for photochromism, clearly differs from the 2 others. (solid black: dark blue; dash black: light blue; dash grey: light blue; solid grey: blueish green; dotted black: purple photochromic)

Sulphur emission.

The orange luminescence, typical for the sodalite group, is attributed to S_2 (sulfide group). It is a **vibronic** spectrum that is caused by S_2^{1-}+ e -> S_2^{2-} . On a broad emission band, you have a number of smaller peaks (that increase in number and sharpness if you cool to liquid nitrogen).

At room temperature, mostly 4-6 vibronic peaks are visible, 20 to 24 nm apart on a broad band between 500 and 800 nm (A Sidike, 2007, Kirk, 1954).

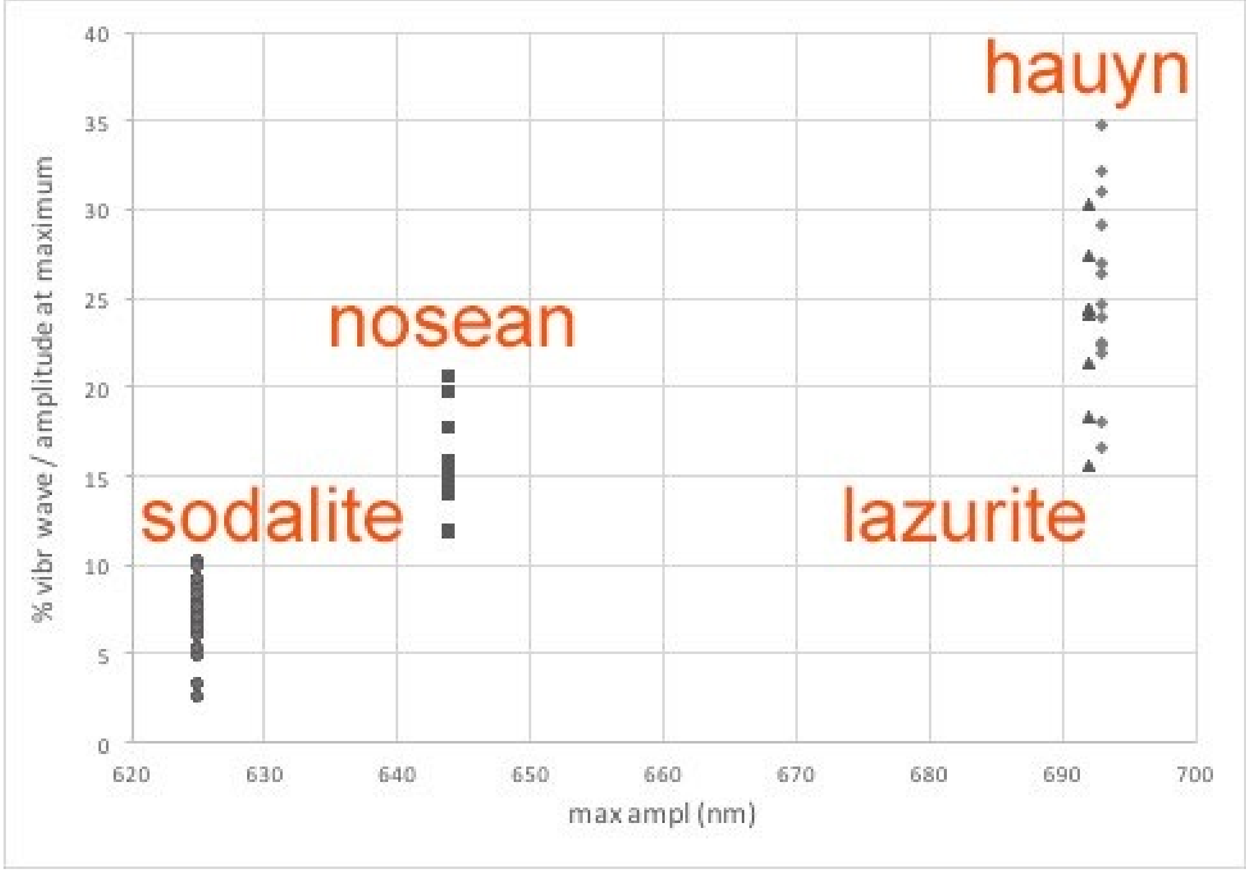


Emission spectra of the 4 minerals (solid black: sodalite; solid grey: lazurite; dashed black: haüyne; dotted black: nosean), corrected for the spectral sensitivity of the spectrometer. Excitation by a blacklight, a 365 nm or a 400 nm UV-A LED

- The sodalite spectrum has the least definition and a maximum around 625 nm. The vibronic ampliture is 7% on average.

- The lazurite/haüyne spectrum has more definition and a maximum around 695 nm. Haüyne has, on average, a larger amplitude vs. lazurite (28% vs 24%).

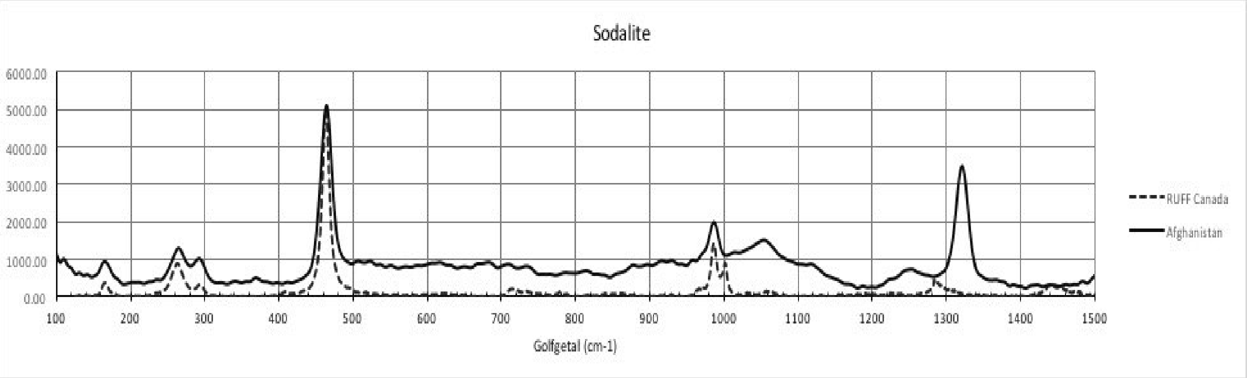
Nosean has a definition between sodalite and haüyne and maximizes around 645 nm. The vibronic amplitude is on average 16%.



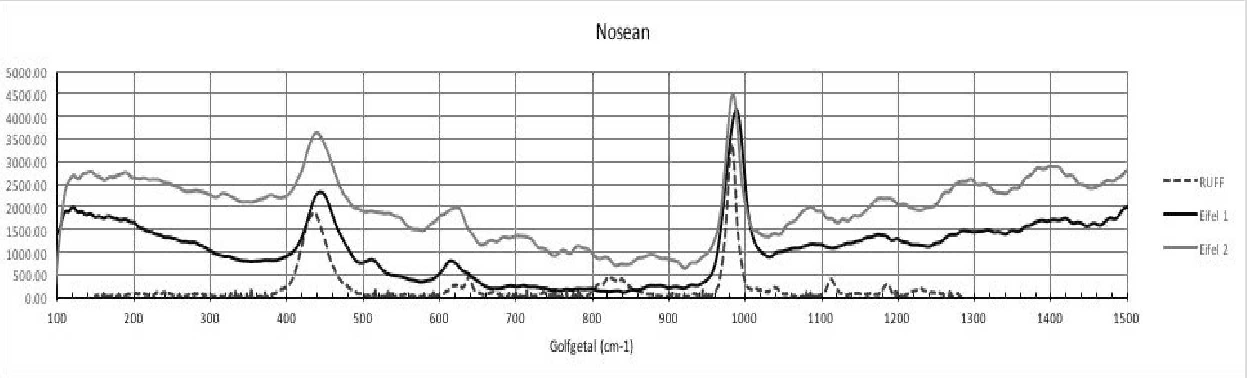
Graph representing the ratio of the vibronic amplitude relative to the continuous emission at the maximum. The solid solution between haüyne and lazurite is clearly visible.The vibronic amplitude is the difference between a maximum and minimum at the maximum envelope. The % is defined as the ratio between the amplitude and the average of the max and min value.

Mineral verification.

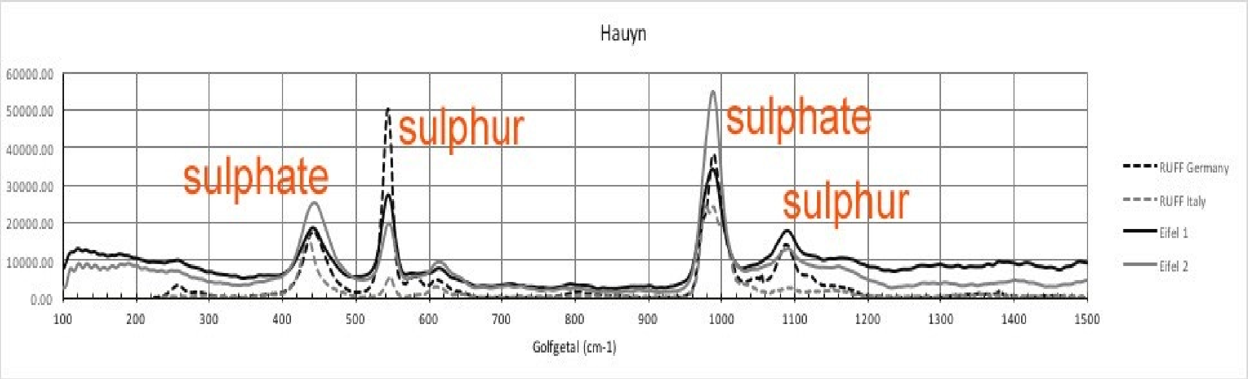
The correct identity of the samples has been checked with Raman and **EDX/SEM**.



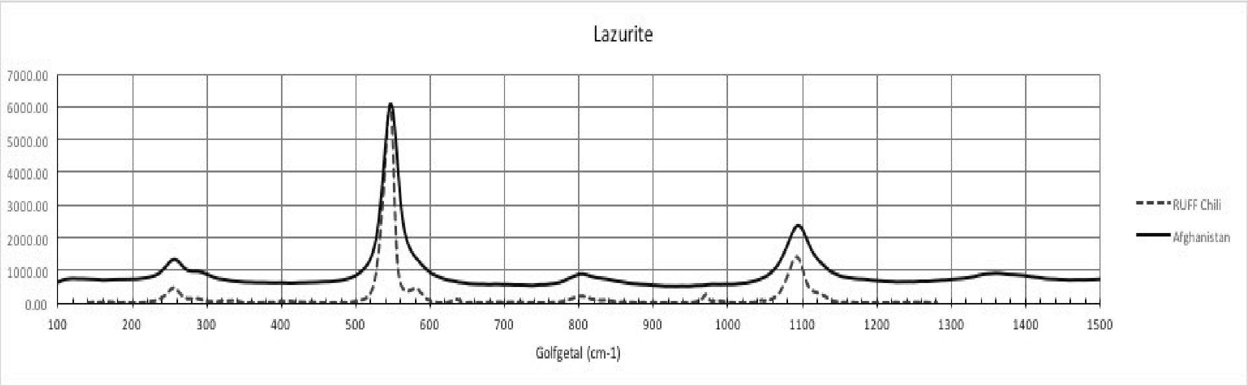
Raman spectrum of a sodalite (identified by its sulfide luminescence) from Koksha, Afghanistan compared to a RUFF sample from Canada.



Raman spectrum of 2 nosean samples (identified by its sulfide luminescence) from the Eifel area, Germany compared to a RUFF sample from the same region.



Raman spectrum of a haüyne samples (identified by its sulfide luminescence) from the Eifel area, Germany compared to a RUFF samples from Germany and Italy. Lines coming from the sulphate group and sulphur clusters are vsisble. Note the difference in ratio between the two line types depending on sample and locality. It demonstrates the solid solution between haüyne and lazurite.



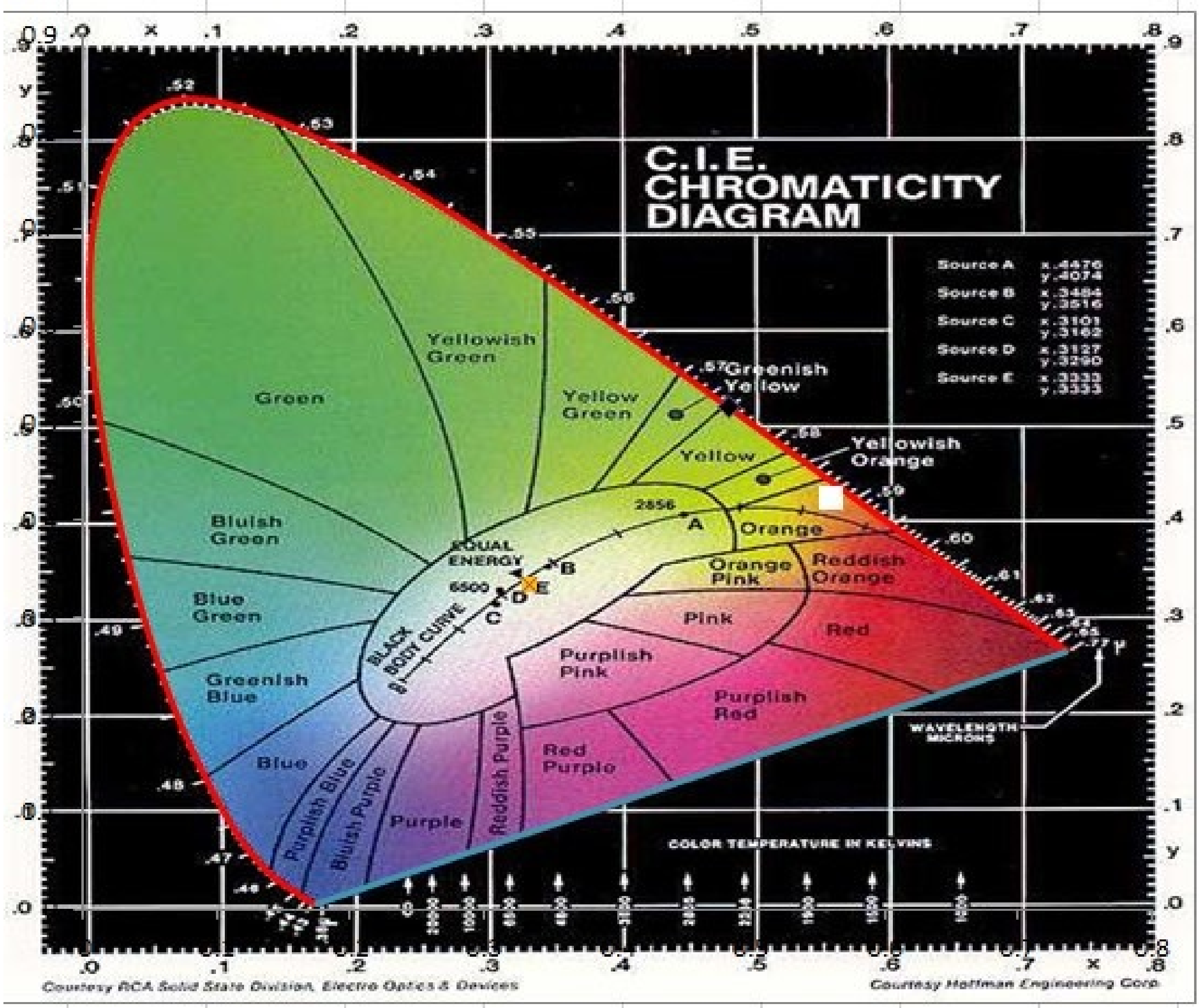
Raman spectrum of a lazurite (identified by its sulfide luminescence) from Koksha, Afghanistan compared to a RUFF sample from Chili. Note that in the chili sample the sulphate peak is not visisble, indicating a sample that is close to the ideal lazurite composition.

Raman spectra were taken at the Ghent University, Chemistry Department, Raman Spectroscopy division (see acknowledgements) on a microscope equipped with a raman spectrometer operating in the green (532 nm) and the red (785 nm).

Emission colour nomenclature.

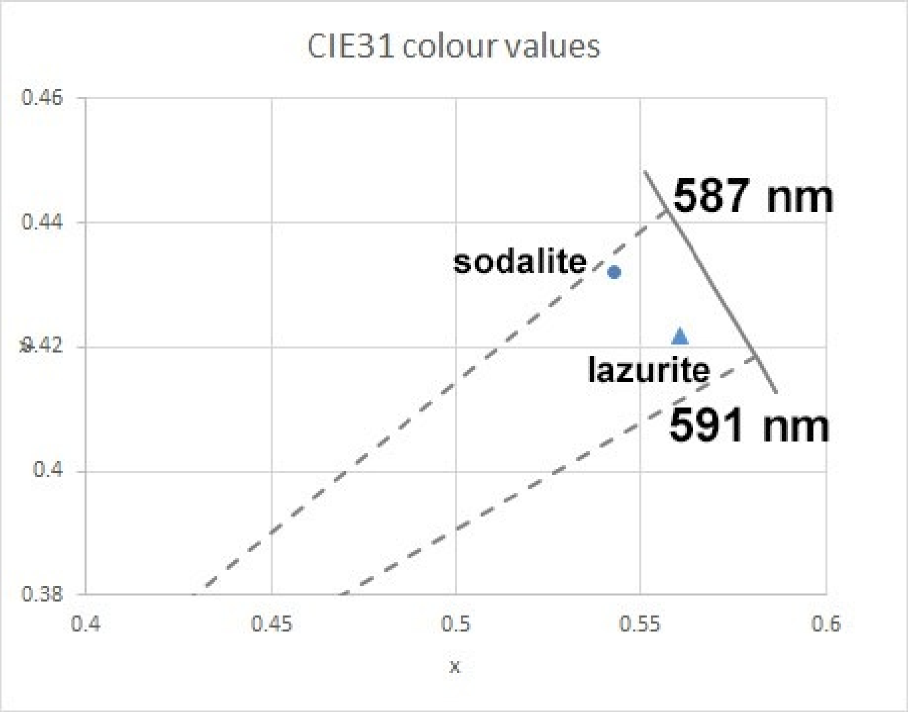
There is always a discussion about naming a colour under mineral collectors. When you have a spectrum, it is possible to calculate colour coordinates, that are universal.

When doing this for the vibronic sulphur emission colour (CIE colour system), you end up with a dominant wavelength of 587 to 591 nm. This gives an official colour name “orange”.



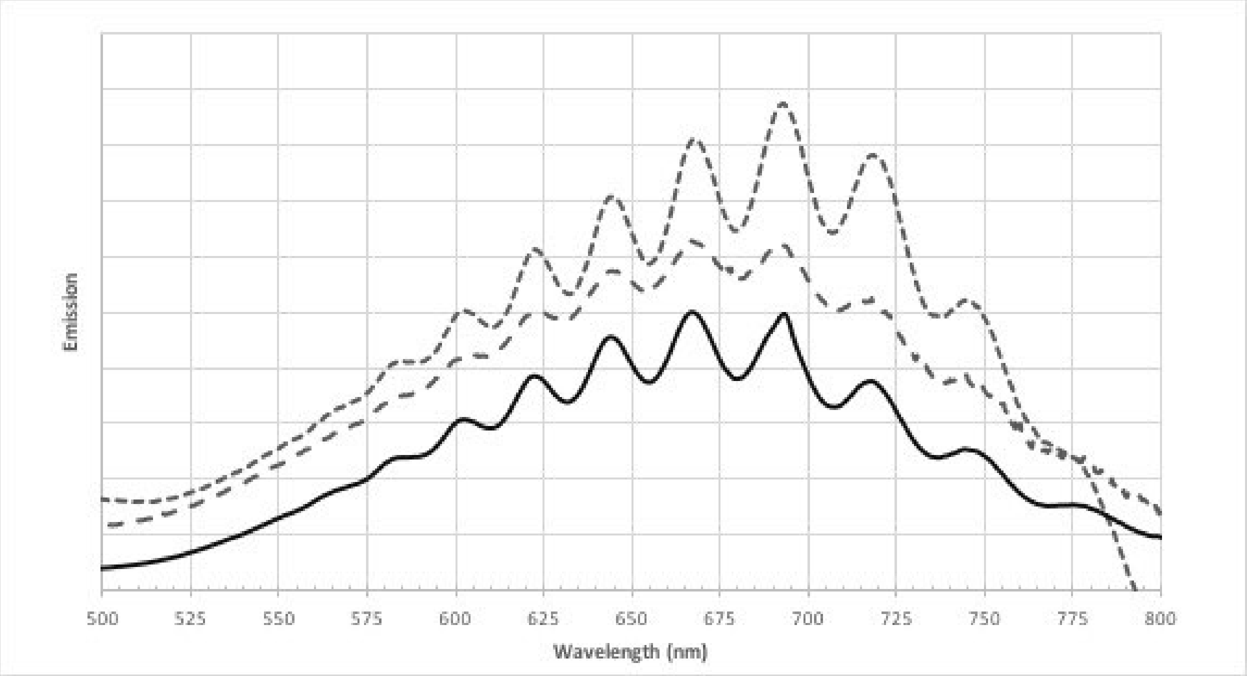
CIE 1931 colour space. The white square is the typical orange colour of the sulfide luminescence in sodalite type minerals.

Since the emission maximum of lazurite/haüyne is more towards the red, the colour coordinate is also slightly red shifted as can be seen in the following enlarged graph of the colour space. Nosean is in between.



The cage problem.

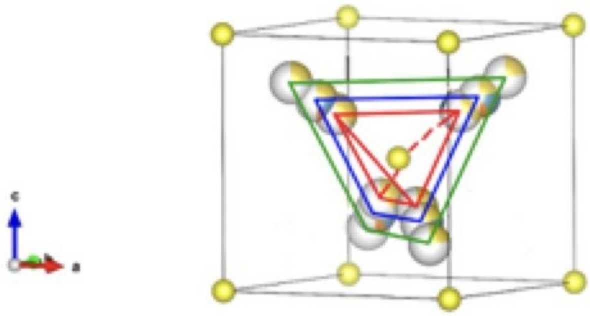
When we look at the vibronic band structure, we don't see a real difference between the 4 minerals (within the accuracy of our spectrometers). The energy difference between the bands is 68 meV (548 cm-1) with a 3-sigma deviation of +/- 5 meV. This is also the energy range we find in the literature for sulfide compounds. For organic molecules a range of 500-700 cm-1 is reported. Measurements on alkali-iodides (Hunsicker,1995) give a value of 600 cm-1.



Emission spectra of haüyne, Eifel (solid), lazurite, Chili (dashed), lazurite, Baikal (dotted)

The spectra of the 4 minerals (beginning of this chapter) show a clear differerence in the place of the maximum and the amplitude of the vibronic bands. The above spectra shows 1 haüyne and 2 lazurites. The 2 lazurites have different amplitudes. These variations occur in all of the 4 minerals. The Chili lazurite only showed sulphur in the Raman spectrum and no sulphate, so it is very sulphur rich (source RUFF). We attribute the smaller amplitude to concentration quenching.

A very plausable explanation is a difference in cage size. Sodalite only has one type of cage and it is small. The three other minerals may have different cages, not only in composition, but also in size.

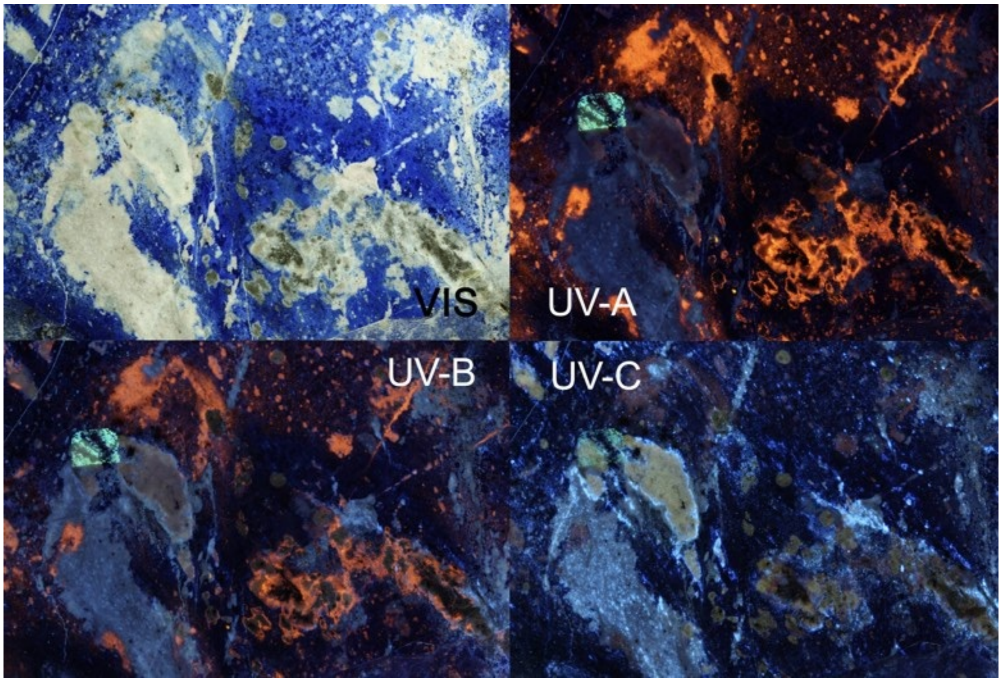


Lazurite, nosean and haüyne have different positions for Na, K and Ca (large spheres) and as such also the cage size. (drawing with VESTA (Momma, 2012)). The silicate lattice is not shown. The smaller spheres represent the anion groups. See e.g. Rastsvetaeva, 2002; and Hassan, 1985 and 1989)

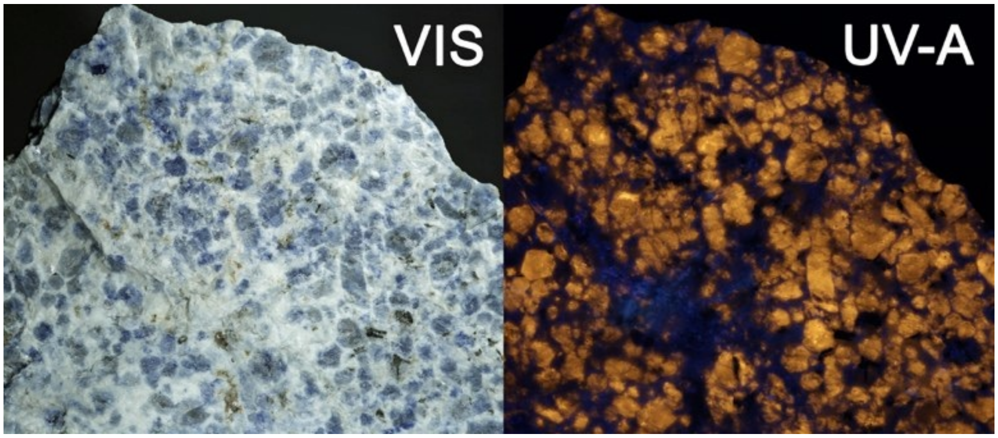
In sodalite the cage is a $[Na_4\ Cl]$ group, in haüyn $[Na_3Ca\ SO_4]$ or $[K_2Ca\ OH]$, in nosean $[Na_4\ SO_4]$ or $[Na_4\ H_2O]$ and in lazurite $[Na_3Ca\ SO_4]$ or $[Na_3Ca\ S_x]$. Probably there exist intermediates because of the solid solutions between the minerals.

Depending on the size of the cage and the interaction with surrounding ions, the sulfide group will be able to vibrate more or less freely, and that will influence the intensity of the bands and the occupation of energy levels and thus the maximum. It is generally assumed that the sulfide group is in the sulphate cages, but in view of the spectral variation, more positions may be possible. A project has been started with the LUMILAB group of the UGent to investigate this further with spectroscopic techniques (see acknowledgements).

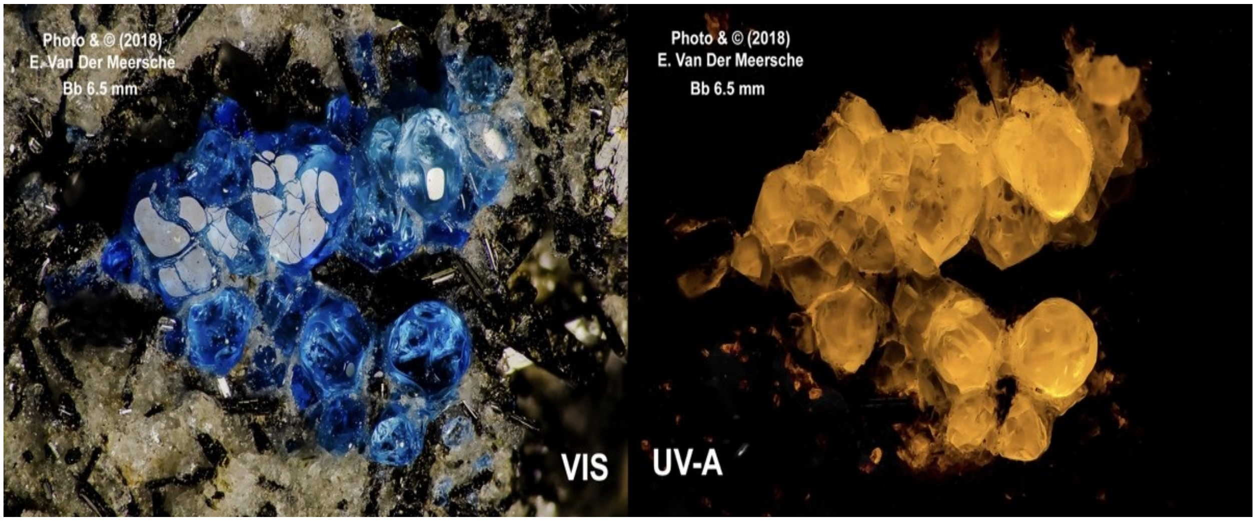
Examples.



Lazurite, Ovalle, Limari, Coquimbo, Chili (RDE). The white UV-C luminescence is probably wollastonite. The locality also reports sodalite, so its not clear whether the blue mineral is sodalite or lazurite. The orange luminescent mineral is lazurite, according to the emission spectrum. UV-A,B,C sources: mercury lamps.

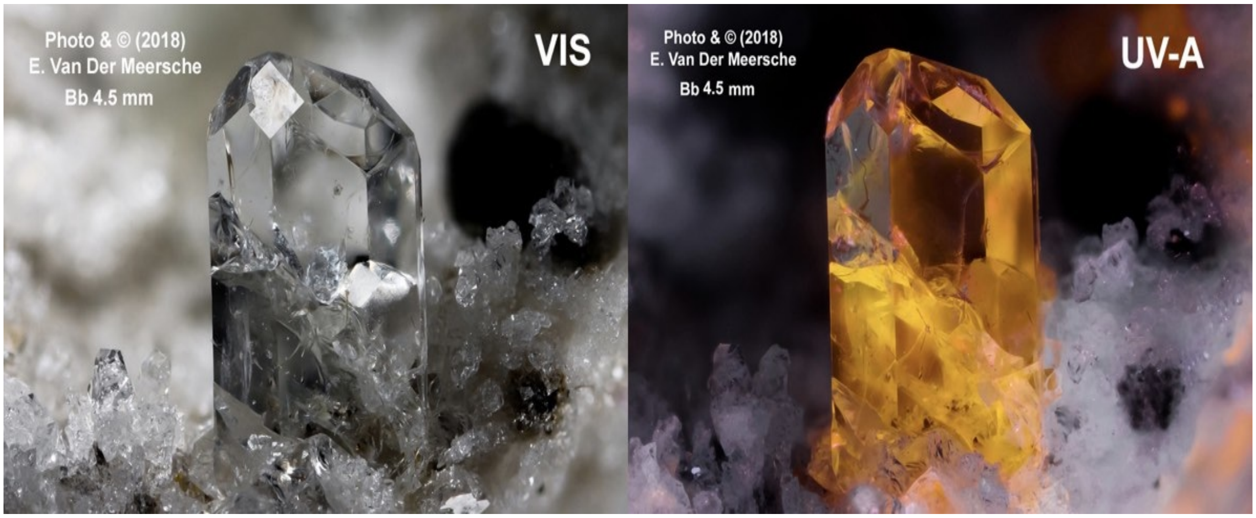


Sodalite, Mont Saint-Hilaire, Quebec, Canada (RDE). It is an alkali rich intrusion that contains sodalite syenite and as such yields larger sodalite crystals. UV-A source: mercury lamp.



Haüyne, Krufter Ofen, Kruft, Eifel, Germany (EVDM and picture). UV-A source: 365 nm LED.

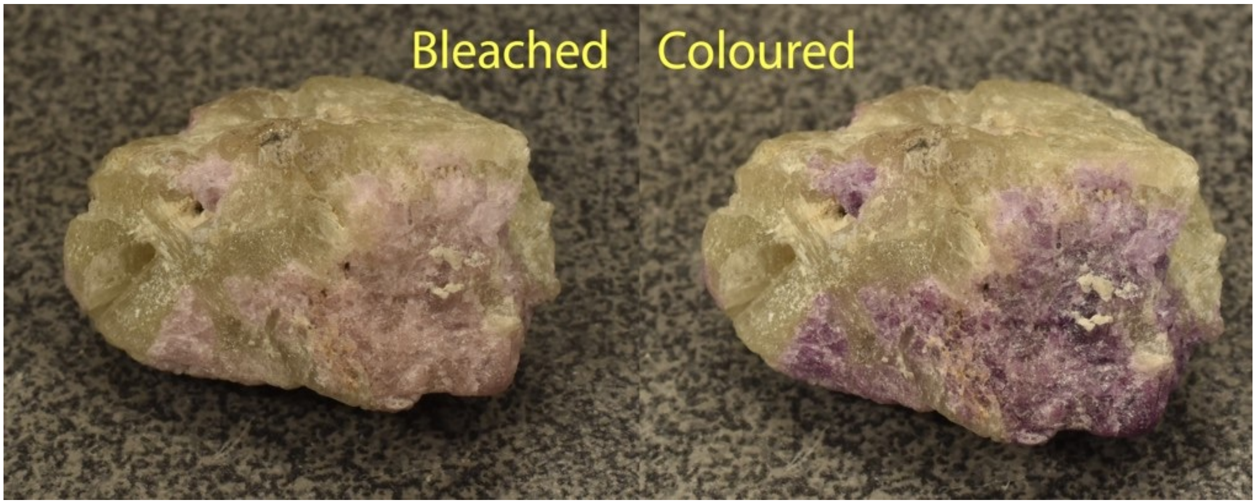
Blue nosean, Laacher Kessel, Mendig, Eifel, Germany (EVDM and picture). UV-A source: 365 nm LED.



Transparent hauyn crystal. UV-A source: 365 nm LED (EVDM and picture).

Photochromic centra.

The photochromic effect in the sodalite group can be seen under the following conditions: Irradiation by UV or blue light (225-480 nm, or even darkness) induces a pink to purple colouration. By using visible light (480-750 nm) the effect is reversed and the sample bleached again (Pizani, 1985). The effect is reversable. High temperature destroys the effect.



Photochromic sodalite before and after irradiation with UV light (Afghanistan, LVG).

The effect is typical for the variety called “hackmanite” (obsolete name) where we choose photochromic sodalite to describe it. When found it will be coloured on a fresh fracture, but bleaches after a while in visible light. In english literature (certainly in the older one) the term “tenebrescent” may be used for the effect.

The effect is linked to **F-centra** (F comes from the German word “Farbe”). F-centra are created when a vacancy from a negative ion is filled with one or more unpaired electrons. They absorb light and this gives colour (Farbe). F-centra in halite are well known and studied. The absorption in sodalite is shown in the reflection spectra in the beginning of this chapter. The first link to an F-center was already done by Medved, 1954.

The models.

What really happens between colouring and bleaching is another story. Everybody agrees that there is an exchange with another center that delivers the electron. From irradiation experiments and consecutive bleaching and heating, several models exist.

Van den Brom (1974) states that the electron comes from Al-tetrahedra groups. They will remain with a hole in one of their bonds. The donated electron then pairs with an extra interstitial sodium-ion that compensates the aluminium group charge. He excludes sulphur as an electron supplier, since the effect is also seen in sulphurless sodalite.

Pizani (1985) Starts with 6 models. After evaluation, he chooses the one that best explains the experimental data. His model for the F-center is an electron captured in the sodium cage where normally one of the anions (chlorine or equivalent) resides, so a [e^- $Na_4{}^+$] group. The electron is coming from a silicon or aluminium oxygen tetrahedron. This should explain two overlapping bands in the spectrum (around 600 and 645 nm). He also excludes sulphur as an electron source.

Finch (2016) gives a global overview of colour centra in sodalite. They studied natural sodalites with different colours. A pink sodalite had its colour coming from the F-center and showed absorption bands at 360 and 530 nm. The blue colour was attributed to a S_3 (tri-sulphur) group. The F-center generates a blue luminescence around 360 nm. Luminescence at 400 and 460 nm is attributed to paramagnetic oxygen defects in analogy to feldspars. It should be noted that they studied X-ray stimulated luminescence and not UV stimulated. They refer to recent literature and state (in contrast to the previous authors) that S_2^- (sulfide) is the electron donator.

Norrbo (2016) states that the F-center is a $[e^- Na_4{}^+]$ complex and that recent literature shows that sulphur donates the electron, most probably in the S_2^- form. They propose a band model that shows the mechanism.

Finally, a theoretical calculation on the F-center (Curutchet, 2016), starting from a trapped electron and S_2^- as electron donor give a spectrum that confirms the observed.

The photochromic effect is also reported in other minerals of the group (Warner, 2012). They studied synthetic hackmanite, sulfo-sodalite and tugtupite.

Other activators.

The literature also deals with the classical metal ions as activators. Three specific recent studies are summarized.

Finch (2016) reports Fe^{3+} with an emission around 680 to 700 nm. Iron replaces aluminium.

Norrbo (2016) made a study of titanium and manganese doped “hackmanite”. They proved that Ti^{4+} is responsable for a blue luminescence around 450 nm and an afterglow peaking at 520 nm. The Mn^{2+} luminescence is seen around 540 nm.

Gaft (2009) identifies Fe^{3+} around 730 nm, Pb^{2+} around 440 nm, Cr^{3+} around 687 Nm and Hg^{2+} around 450 nm.

General information on possible activators can be found in Gaft (2005 and 2015), 2 references that are the successor of the previous work of Gorobets and Rogojine (2002).

CHAPTER 7

The Koksha Minerals

We received a large number of samples from the Koksha area for our study from Marc Cole and Rob Woodside (see acknowledments). Since you find many minerals from the sodalite group, an ideal test case to distinguish among them. Furthermore, the specimen (Herwig Pelckmans) with the special luminescence and afterglow (see introduction) also came from this area.

The Koksha area.

The Koksha area, also known as Sar-e-Sang, lies in the Badakshan Province in Afghanistan. Sar-e-Sang is just one of the localities in the valley formed by the Koksha river. A very good description of the area and their minerals can e.g. be found in J. Wyart (1981) and T. Moore (2014).

Badakhshan is a province in the north-east of Afghanistan, sandwiched between Pakistan and Tadzjikistan. The Koksha valley lies in the Hindu Kush massif, having many peaks above 6000 m. Sar-e-Sang lies around 2500m altitude. To reach it you need a 4x4 and mules.

The area is strongly metamorphised and contains gneiss, schists, amphibolites, pyroxenites and many other rock types. Biggest problem is the correct location of your specimen. In the whole area Mindat mentions 3 zones containing 15 localities. Since all minerals are transported to collection points, a correct localisation is difficult.

The minerals and their identification.

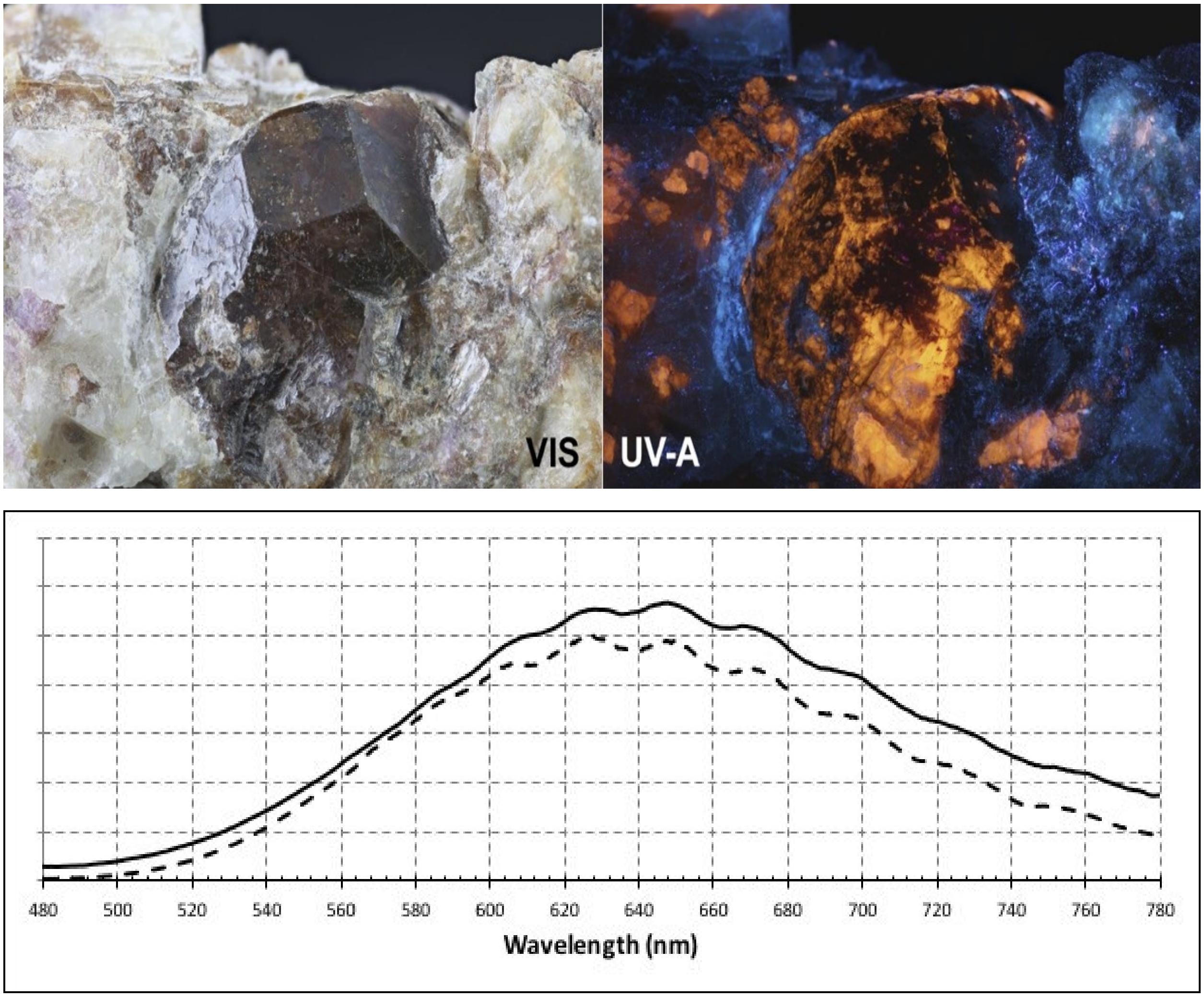
The minerals received from our 2 contacts all had a label. By using the luminescence we were able to check the correctness.

Besides the sodalite group minerals, there were also 2 interfering minerals: marialite (the sodium scapolite) and afghanite (cancrinite like structure). Both are very related structurewise to sodalites and also exhibit a vibronic sulfide spectrum. They will be the subject of a seperate study.

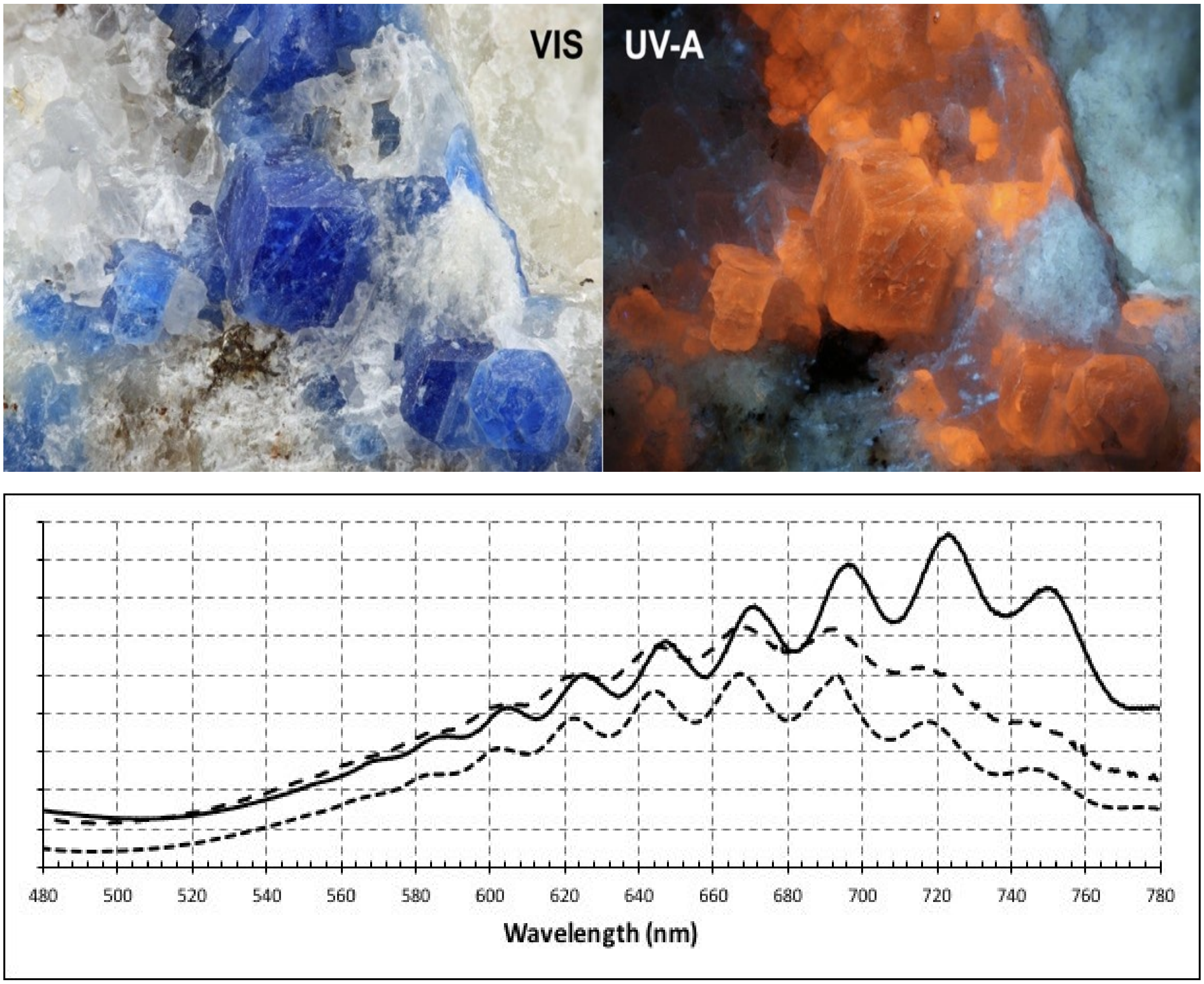
By using reference material, we could determine they both have a different maximum and can be distinguished from the sodalite series.

Besides these, there was also blueish white luminescing gonnardite that occurs as a transformation product of lazurite.

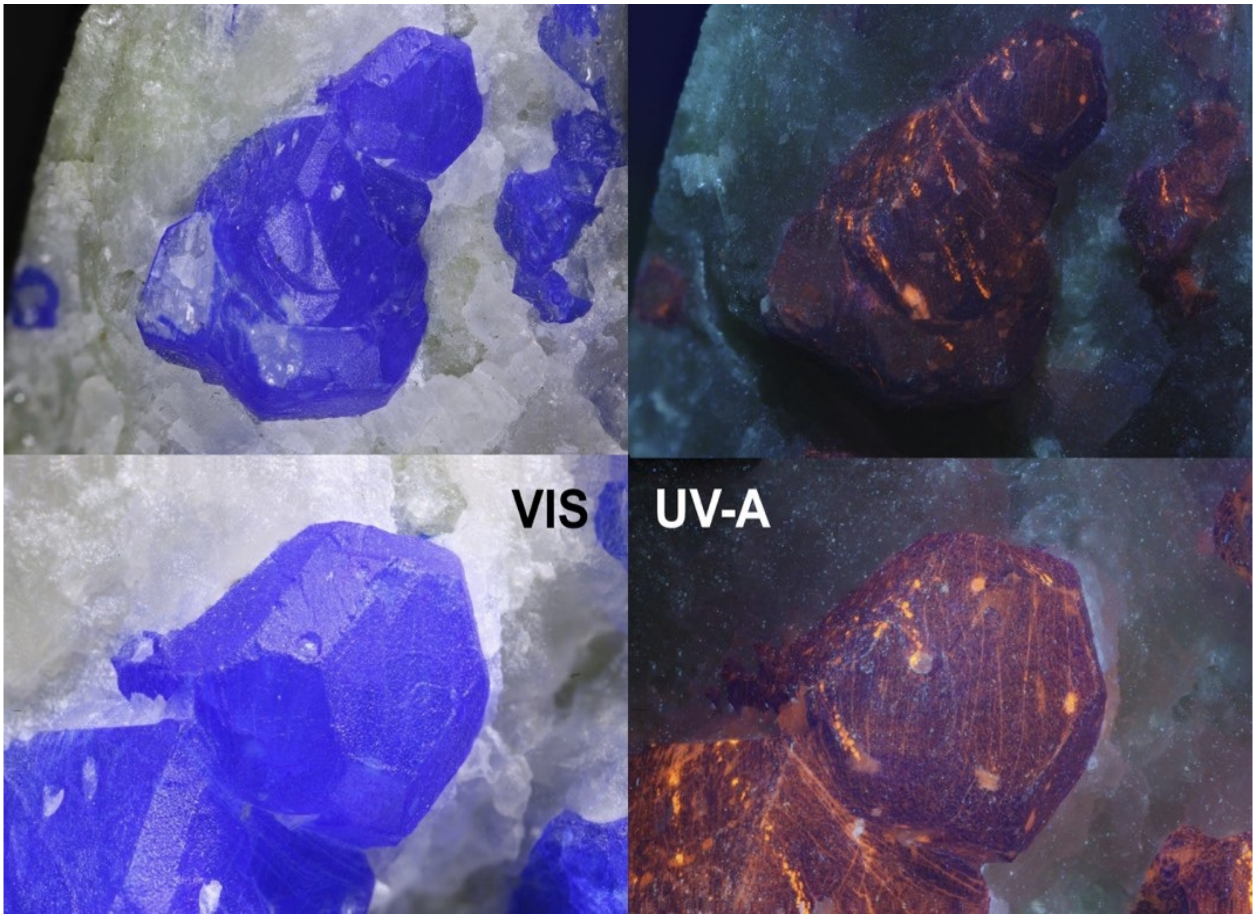
In the following pages we will give some examples of the analysis.

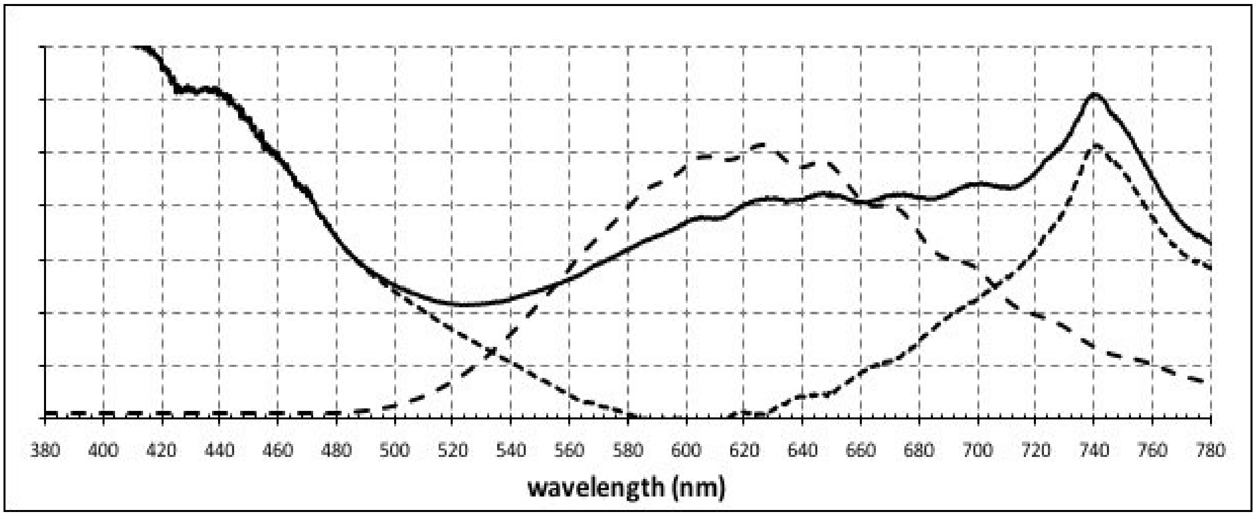


The brown dodecahedron clearly has a sodalite spectrum (dashed = reference) (AEM, MCO). UV-source: UV-A mercury lamp and 365 nm LED

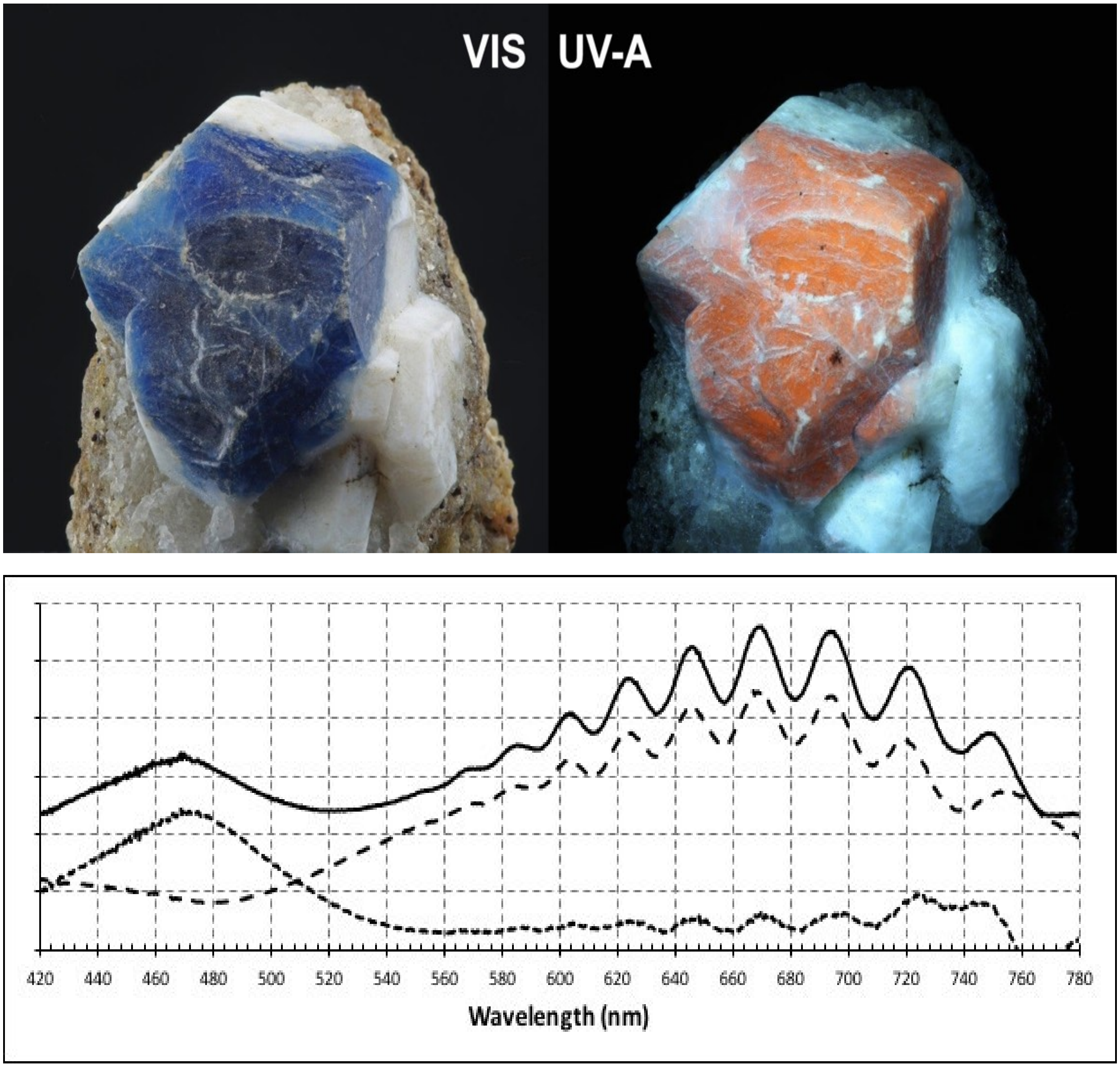


The blue crystals clearly have a lazurite/haüyne spectrum (dashed ref: lazurite Chili, dotted ref: haüyne Germany). The fact that the maximum is shifted may be due to an underlying luminescence (see further examples) (AEM/RWO). UV-source: UV-A mercury lamp and 365 nm LED.





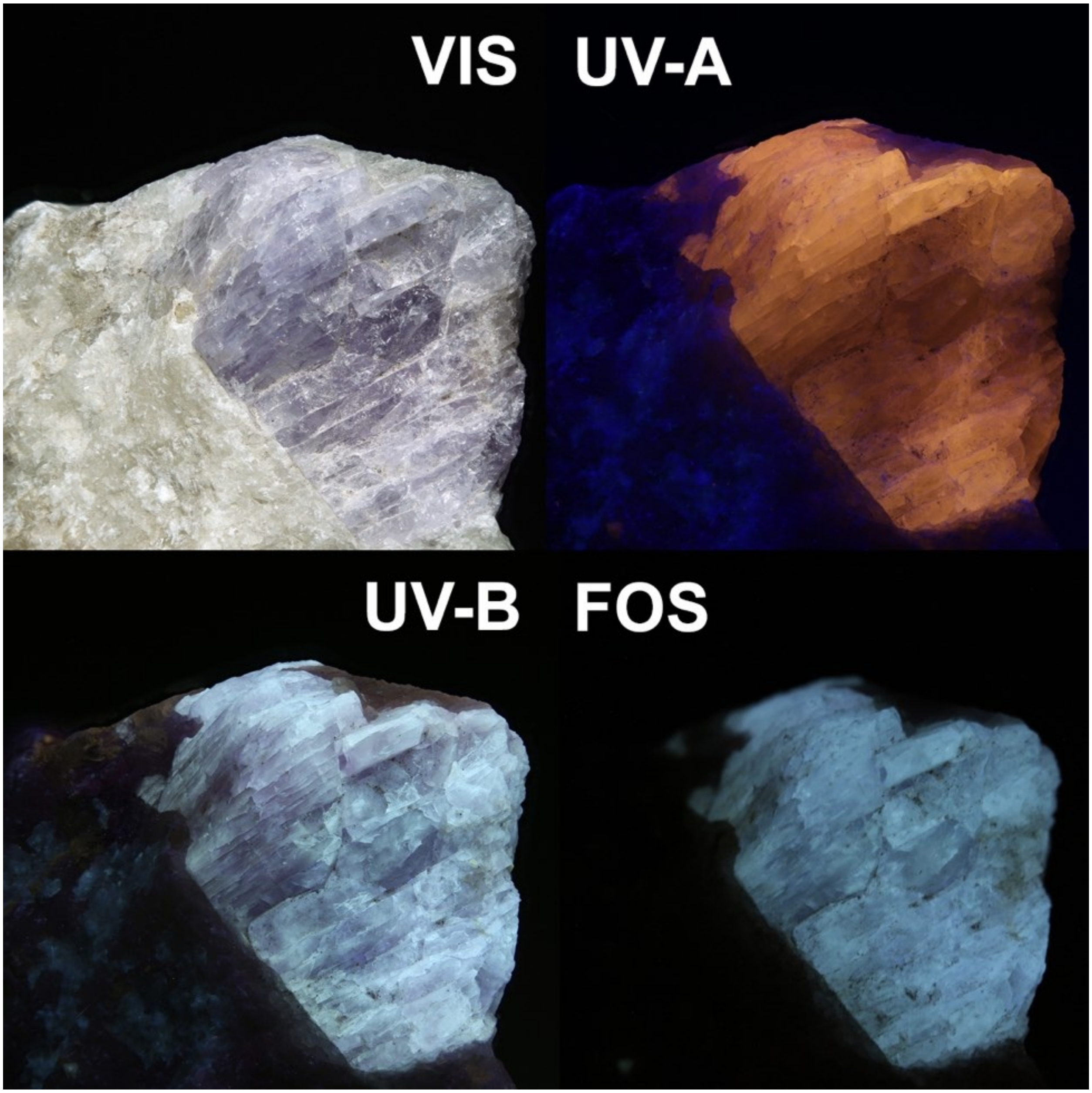
The specimen clearly has a sodalite spectrum (dashed = ref). After removal of the sulfide part (dotted), we see an emission in the red (740 nm) of Fe3+ and an emission in the green/blue, probably Ti or lattice defects (AEM/RWO) UV-source: UV-A mercury lamp and 365 nm LED.



This specimen is clearly lazurite (dashed = ref), but after subtraction we see a contribution of Fe^{3+} *at 740 nm and an emission in the green at 470 nm (probably* Mn^{2+}*). The fact that the vibronic structure is not completely removed may point to a slightly different cage size relative to the reference. (AEM/RWO). UV-source: UV-A mercury lamp and 365 nm LED.*

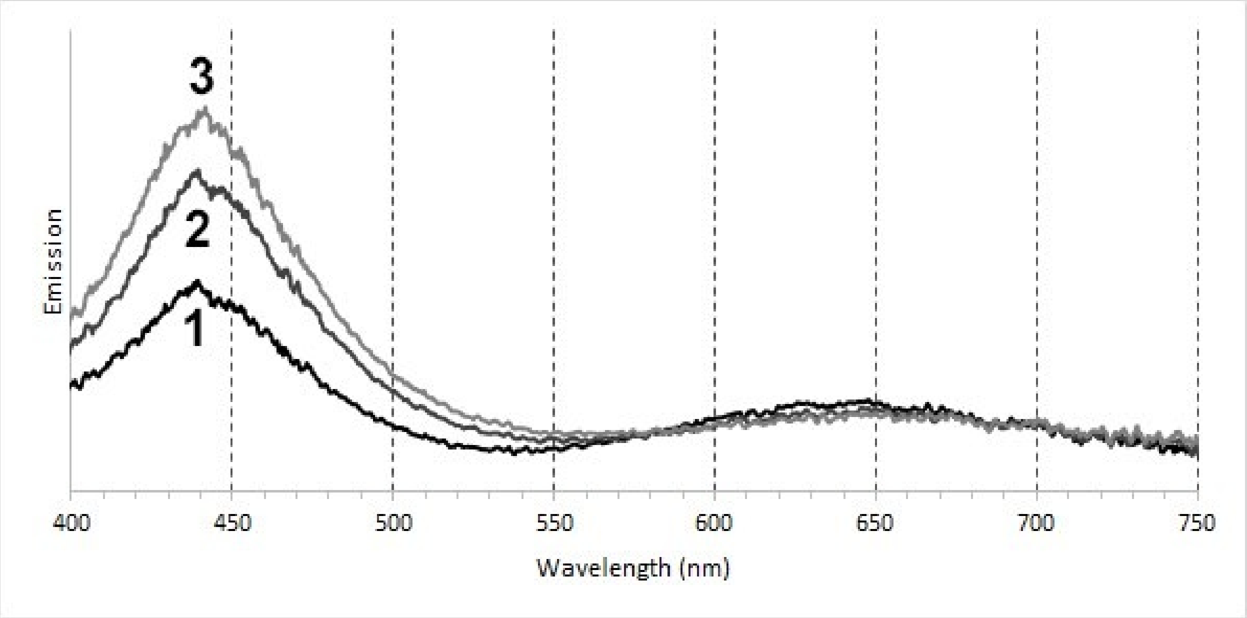
White luminescence and afterglow.

The “special” HPE sample referred to in the introduction proved not to be alone and not to be limited to the Koksha area.

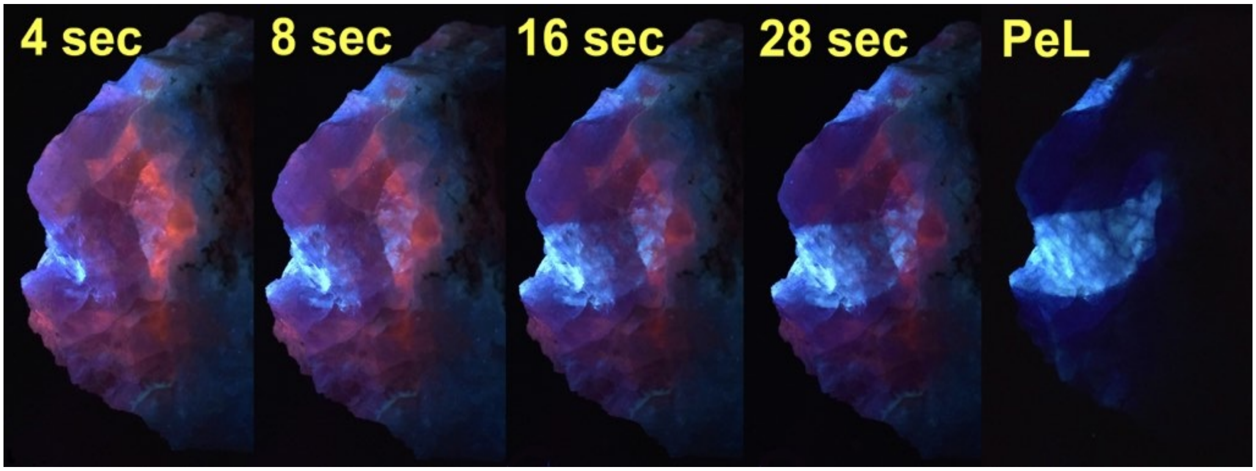


Photochromic sodalite showing an orange sulfide luminescence under UV-A. Under UV-B, the resulting luminescence is blueish white (also under UV-C). A blueish white afterglow (FOS) remains after switching of the UV-B (HPE).

The UV-B luminescence often starts with orange, but is then shifted towards blueish white. Measurements on a slower reacting sample showed that the orange luminescence does not disappear, but gets dominated by the rising blueish white luminescence. A weak vibronic sodalite structure is visible in the orange luminescence. When switching off the UV, a white phophorescence remains.



Emission spectrum of a typical sample. The orange luminescence stays stable, but the blue emission rises in time (1: at start, 2 and 3 subsequent time intervals). UV-source: UV-B LED 310 nm. Below a photographic sequence (4 sec exposure intervals). The dimming of the orange is due to to the increase of the purple colouration.



By using the website of Marc Cole that contains a large number of data on luminescent minerals, we did the following observations:

The blue luminescence and afterglow is rare but not linked to Afghanistan alone. You also see it in Canadian sodalite and Greenland one.

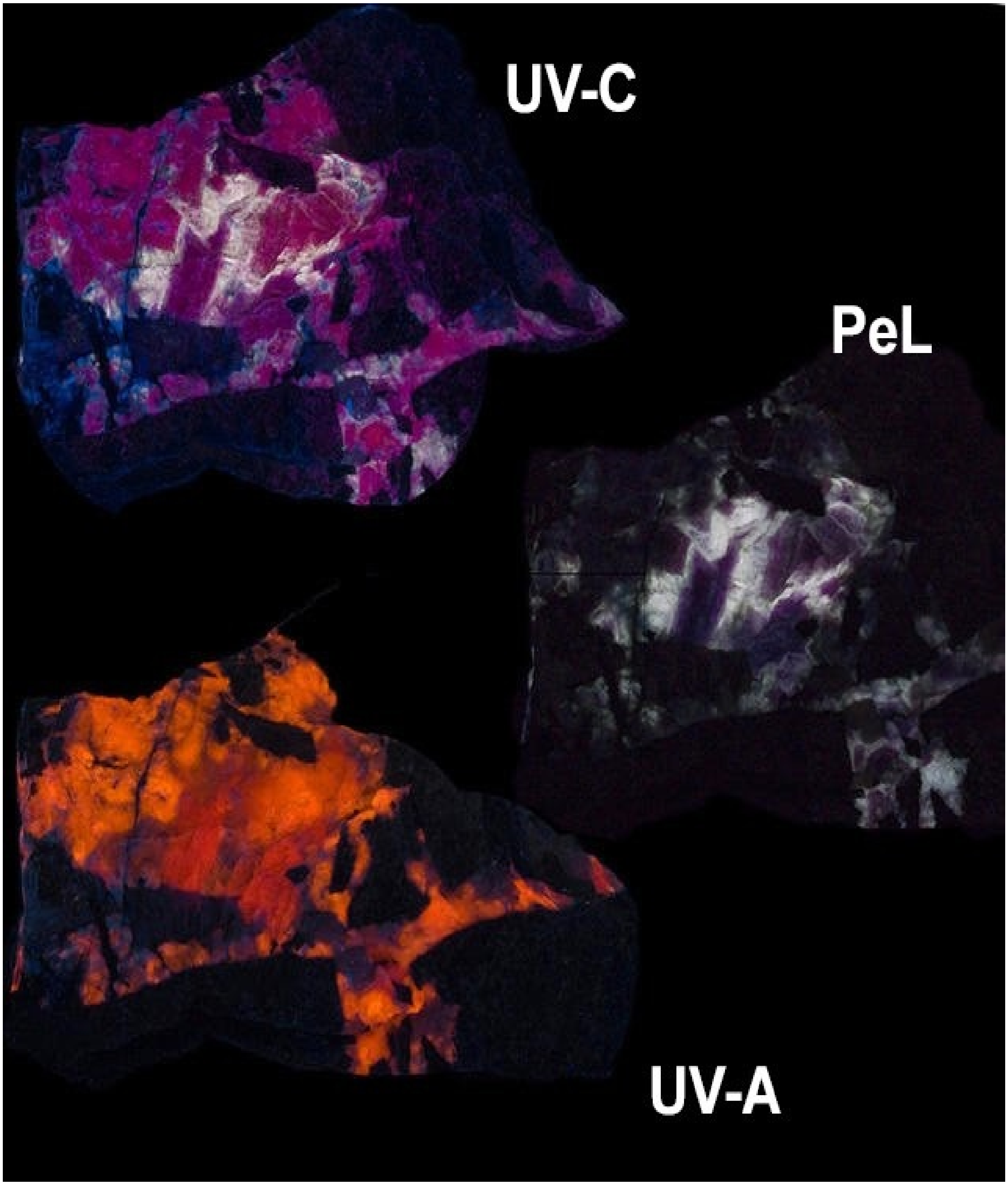
- You only see it in photochromic sodalite that is white luminescent. Photochromic sodalite with a red Fe^{3+} UV-C luminescence does not show the effect.

- Pieces showing the white luminescence and afterglow are also thermoluminescent.

In every locality, we found an example that contains zones having a red, non-phosphorescent and a white luminescent and phosphorescent part.

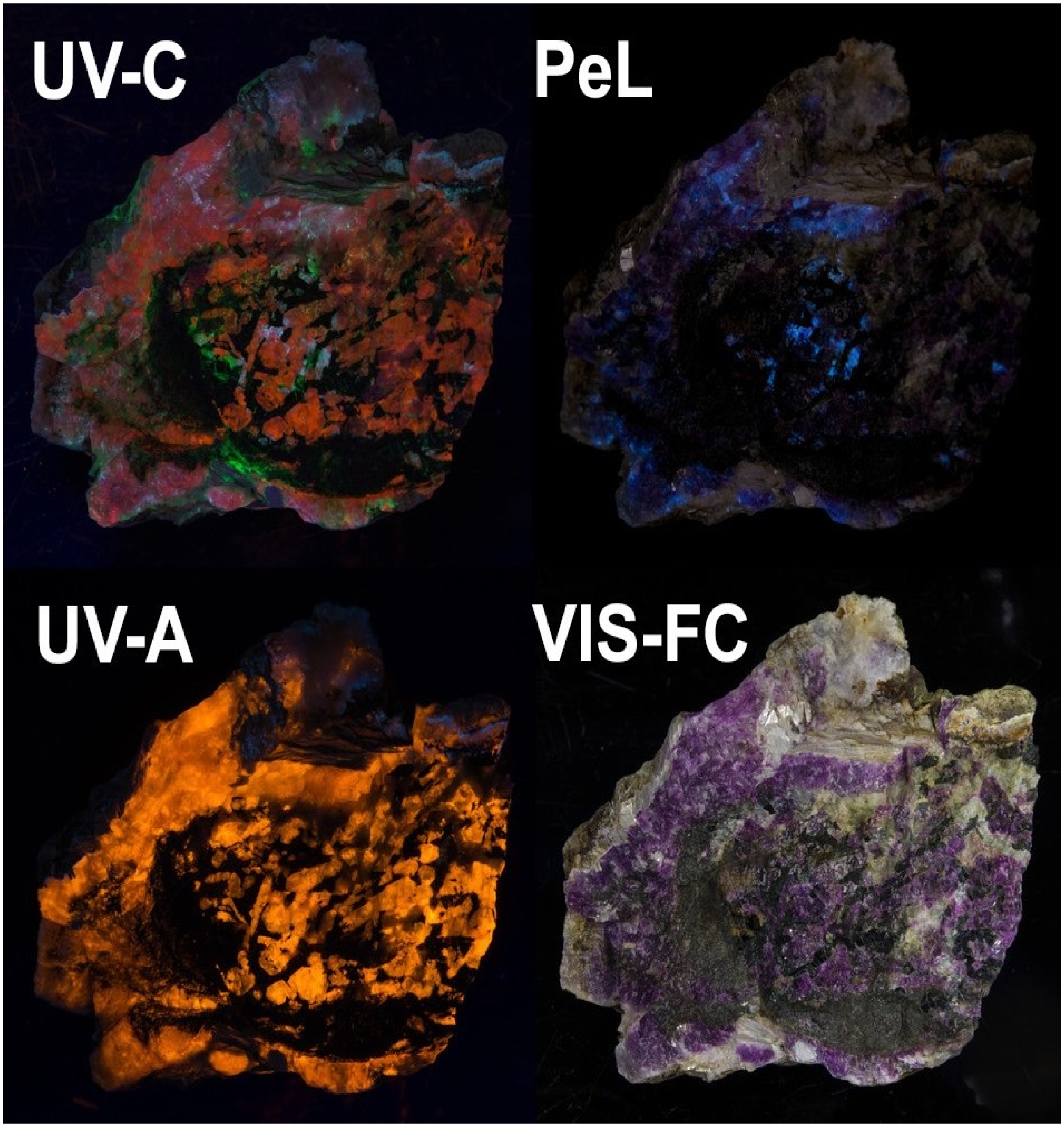
Titanium is proposed as the activator for the white luminescence. We provided samples to the Chemistry department of the Turku University, Finland who already published on this subject (Norrbo, 2016). An new article on the mechanism is under preparation (Lastusaari, private communication, 2018). The effect is also described by Wagner, 2012.

The three referred specimens from the nature’s rainbows website with mixed zones are shown in the following pages:

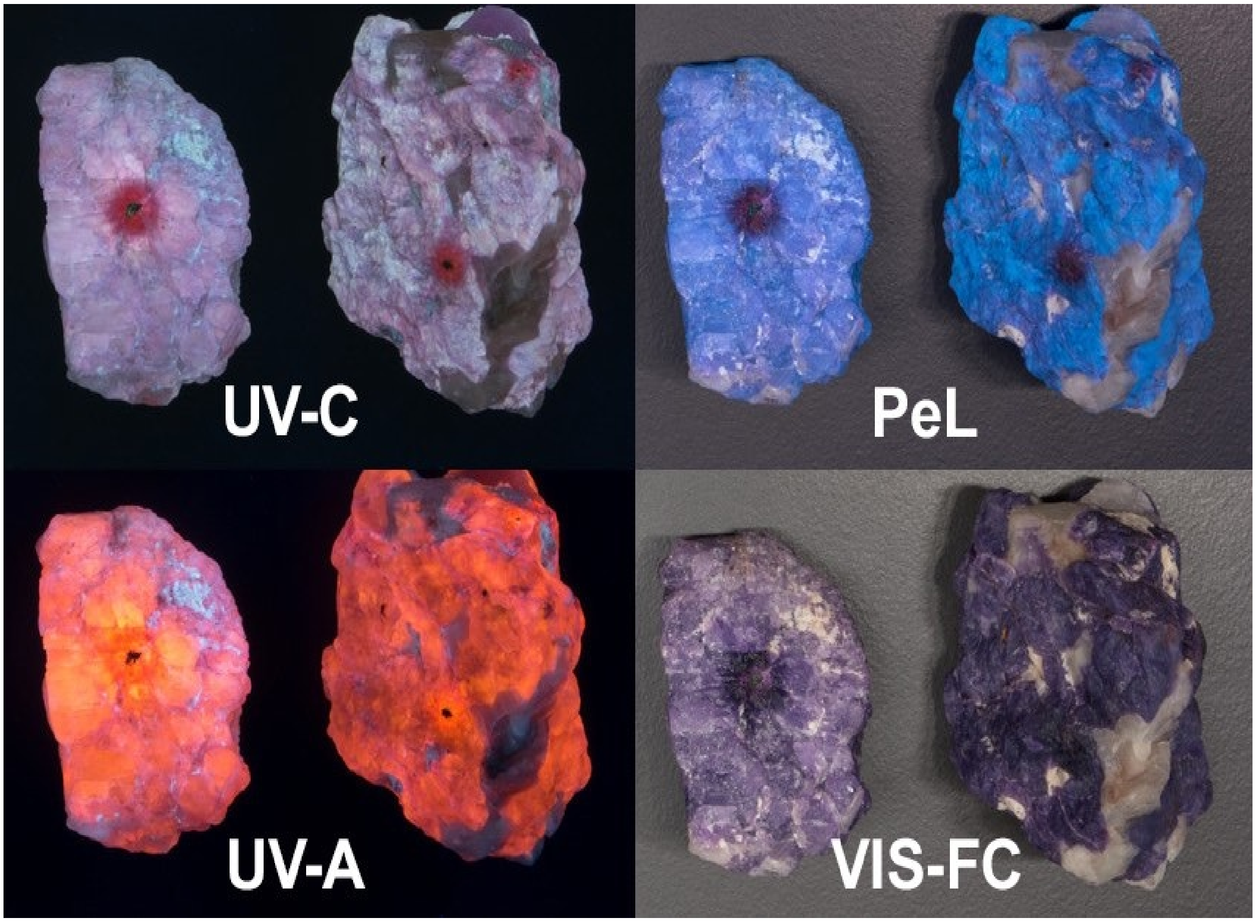


Mt-St-Hilaire, Quebec, Canada. (PeL stands for persistent luminescence, another term for afterglow)

https://www.naturesrainbows.com/single-post/2015/12/01/Sodalite-Hackmanite-Mt-St-Hilaire-Canada



Ilimausaq complex, Greenland: *https://www.naturesrainbows.com/single-post/2016/09/29/Sodalite-and-Ussingite-w-Epistolite---Ilimaussaq-Complex-Greenland*



Sar-e-Sang, Koksha valley, Afghanistan: *https://www.naturesrainbows.com/single-post/2016/11/22/Purple-Hackmanite---Afghanistan*

CHAPTER 8

Spectrometers, UV-Sources

The following spectrometers have been used:

Ocean Optics Jaz spectrometer 180 - 1080 nm

Ocean Optics Flame spectrometer 200 - 1100 nm

Ocean Optics USB 2000+ 190 – 888 nm

The following UV light sources were used :

UV-A: Blacklight mercury lamp (centered around 365 nm), LEDs 365, 400 nm

UV-B: LED 305 nm and 310 nm

UV-C: LED 255 nm and 265 nm

The different UV ranges are defined in ISO 21348, included in EEC directive 2006/25/EC and published by Belgian legislation in KB dd 01/02/2010 (For other countries, check your local legislation gazette). The limits are as follows:

UV-A: 315-400 nm

UV-B: 280-315 nm

UV-C: 100-280 nm

The old terminology longwave, midwave and shortwave are not official and were originally used to distinguish between the three types of existing mercury lamps (365nm, 302nm and 254nm). Certainly nowadays with the variety of LED's it is better to specify the correct UV-area and specify the used wavelength.

The spectra in this article were corrected for the spectral sensitivity of the spectrometer. This is done by using a halogen lamp with a known colour temperature. The easy and cheap method fits the recorded spectrum to a black body with the same colour temperature.

A more complicated and more expensive method uses a calibrated halogen lamp. Advantage of this method is that the energy output is known. When using the correct integration of the luminescence signal, an intensity in energy units can then be given.

We used both methods and got analogue results within the accuracy of the different methods. This means that the cheap method can be used by everyone who does not want to invest in an expensive calibrated lamp. In the UV and violet, the deviation is larger, but this has to do with the lower light intensity of the lamps inducing a higher uncertainty in the measurement. The deviation is however only in intensity and general shape and does not influence conclusions.

Use of the calibrated lamp allows to use the internal software of Ocean Optics and requires less fundamental knowledge of the user, but it implies also less flexibility for personal calculations that are not standard.

CHAPTER 9

Luminescence Terminology

We will use the following terminology which is nowadays accepted in scientific environments.

luminescence: also called fluorescence if the decay time is so short (ns to us) that the effect stops (for the human eye) if the exciting light source is switched off. Example: rare earths in scheelite.

phosphorescence: the decay time is longer (us to ms) and after switching off the light source a short flash can still be seen. Example: the red Mn^{2+} luminescence in calcite. the luminescence is driven by forbidden transitions that become allowed due to symmetry breaking.

afterglow: also called long lasting phosphorescence or persisted luminescence when the decay time is so long that an afterglow of seconds to hours is seen after switching of the light source. The luminescence is trap driven here. Example: the greenish yellow organic luminescence in calcite. It is afterglow that most collectors call phosphorescence.

thermoluminescence: is actually the same process as afterglow. To release the electrons out of a trap (that is why they call it a trap), you need a certain amount of energy. If you see afterglow, it means that room temperature energy is sufficient to do that. If you have to heat the sample to see it, it means you need higher energies to kick them out.

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Photochromic

A photochromic substance exhibits photochromism. The ability to change colour upon exposure to electromagnetic radiation.

See also: https://en.wikipedia.org/wiki/Photochromism

Luminescence

Luminescence in this book is the spontaneous emission of light after stimulation by electromagnetic energy.

See also: https://en.wikipedia.org/wiki/Luminescence

Spectrometers

A spectrometer (/spɛkˈtrɒmɪtər/) is a scientific instrument used to separate and measure spectral components of a physical phenomenon. Spectrometer is a broad term often used to describe instruments that measure a continuous variable of a phenomenon where the spectral components are somehow mixed. In visible light a spectrometer can for instance separate white light and measure individual narrow bands of color, called a spectrum.

See also: https://en.wikipedia.org/wiki/Spectrometer

Ultramarine

Ultramarine is a deep blue color and a pigment which was originally made by grinding lapis lazuli into a powder.[1] The name comes from the Latin ultramarinus, literally "beyond the sea", because the pigment was imported into Europe from mines in Afghanistan by Italian traders during the 14th and 15th centuries.

See also: https://en.wikipedia.org/wiki/Ultramarine

Syenite

Syenite is a coarse-grained intrusive igneous rock with a general composition similar to that of granite, but deficient in quartz, which, if present at all, occurs in relatively small concentrations (<5%). Some syenites contain larger proportions of mafic components and smaller amounts of felsic material than most granites; those are classed as being of intermediate composition. The volcanic equivalent of syenite is trachyte.

See also: https://en.wikipedia.org/wiki/Syenite

Silicate

In chemistry, a silicate is any member of a family of anions consisting of silicon and oxygen. Silicates are extremely important materials, both natural (such as granite, gravel, and garnet) and artificial (such as Portland cement, ceramics, glass, and waterglass), for all sorts of technological and artistic activities.

See also:

https://en.wikipedia.org/wiki/Silicate

http://www.galleries.com/Silicates

http://www.science.uwaterloo.ca/~cchieh/cact/applychem/silicate.html

https://www.britannica.com/science/silicate-mineral

Zeolite

Zeolites are microporous, aluminosilicate minerals commonly used as commercial adsorbents and catalysts.[1] The term zeolite was originally coined in 1756 by Swedish mineralogist Axel Fredrik Cronstedt, who observed that rapidly heating the material, believed to have been stilbite, produced large amounts of steam from water that had been adsorbed by the material. Based on this, he called the material zeolite, from the Greek ζέω (zéō), meaning "to boil" and λίθος (líthos), meaning "stone"

See also:

https://www.mindat.org/min-4395.html

https://en.wikipedia.org/wiki/Zeolite

https://www.britannica.com/science/zeolite

Point group

In geometry, a point group is a group of geometric symmetries (isometries) that keep at least one point fixed.

See also:

https://en.wikipedia.org/wiki/Point_group

https://www.staff.ncl.ac.uk/j.p.goss/symmetry/index.html

Specific -4 3 m: https://www.staff.ncl.ac.uk/j.p.goss/symmetry/Td.html

Space group

In mathematics, physics and chemistry, a space group is the symmetry group of a configuration in space, usually in three dimensions.[1] In three dimensions, there are 219 distinct types, or 230 if chiral copies are considered distinct.

See also:

https://en.wikipedia.org/wiki/Space_group

https://en.wikipedia.org/wiki/List_of_space_groups

http://img.chem.ucl.ac.uk/sgp/mainmenu.htm

Lattice parameter

The lattice constant, or lattice parameter, refers to the physical dimension of unit cells in a crystal lattice. Lattices in three dimensions generally have three lattice constants, referred to as a, b, and c. However, in the special case of cubic crystal structures, all of the constants are equal and we only refer to a. Similarly, in hexagonal crystal structures, the a and b constants are equal, and we only refer to the a and c constants. A group of lattice constants could be referred to as lattice parameters. However, the full set of lattice parameters consist of the three lattice constants and the three angles between them.

See also: https://en.wikipedia.org/wiki/Lattice_constant

Dodecahedral

In geometry, a dodecahedron (Greek δωδεκάεδρον, from δώδεκα dōdeka "twelve" + ἕδρα hédra "base", "seat" or "face") is any polyhedron with twelve flat faces.

See also: https://en.wikipedia.org/wiki/Dodecahedron

Twinning

Crystal twinning occurs when two separate crystals share some of the same crystal lattice points in a symmetrical manner. The result is an intergrowth of two separate crystals in a variety of specific configurations. The surface along which the lattice points are shared in twinned crystals is called a composition surface or twin plane.

See also: https://en.wikipedia.org/wiki/Crystal_twinning

Plutonic rocks

Among igneous rocks, i.e. those formed from magma or molten rock, the most important difference may be between plutonic and volcanic rocks. Plutonic rocks are formed underground.

See also:

http://www.friesian.com/pluton.htm

https://study.com/academy/lesson/volcanic-vs-plutonic-igneous-rocks-definition-and-differences.html

Pegmatite

A pegmatite is an igneous rock, formed underground, with interlocking crystals usually larger than 2.5 cm in size (1 in). Most pegmatites are found in sheets of rock (dikes and veins) near large masses of igneous rocks called batholiths.

See also:

https://en.wikipedia.org/wiki/Pegmatite

https://geology.com/rocks/pegmatite.shtml

Metamorphic

Metamorphic rocks arise from the transformation of existing rock types, in a process called metamorphism, which means "change in form".[1] The original rock (protolith) is subjected to heat (temperatures greater than 150 to 200 °C) and pressure (100 megapascals (1,000 bar) or more), causing profound physical or chemical change. The protolith may be a sedimentary, igneous, or existing metamorphic rock.

See also:

https://en.wikipedia.org/wiki/Metamorphic_rock

https://geology.com/rocks/metamorphic-rocks.shtml

Phonolitic

Phonolite is an uncommon volcanic rock, of intermediate chemical composition between felsic and mafic, with texture ranging from aphanitic (fine-grain) to porphyritic (mixed fine- and coarse-grain).

The name phonolite comes from the Ancient Greek meaning "sounding stone" because of the metallic sound it produces if an unfractured plate is hit; hence the English name clinkstone.

See also: https://en.wikipedia.org/wiki/Phonolite

Raman spectroscopy

Raman spectroscopy (/ˈrɑːmən/; named after Indian physicist Sir C. V. Raman) is a spectroscopic technique used to observe vibrational, rotational, and other low-frequency modes in a system.[1] Raman spectroscopy is commonly used in chemistry to provide a structural fingerprint by which molecules can be identified.

See also:

https://en.wikipedia.org/wiki/Raman_spectroscopy

https://www.nanophoton.net/raman/raman-spectroscopy.html

https://www.youtube.com/watch?v=SsIYDEma_cU

Vibronic

Vibronic spectra involve simultaneous changes in the vibrational and electronic energy states of a molecule.

See also: https://en.wikipedia.org/wiki/Vibronic_spectroscopy

EDX/SEM

Energy-dispersive X-ray spectroscopy (EDS, EDX, EDXS or XEDS), sometimes called energy dispersive X-ray analysis (EDXA) or energy dispersive X-ray microanalysis (EDXMA), is an analytical technique used for the elemental analysis or chemical characterization of a sample. It relies on an interaction of some source of X-ray excitation and a sample.

See also:

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https://en.wikipedia.org/wiki/Scanning_electron_microscope

F-centra

An F-center, Farbe center or color center (from the original German Farbzentrum; Farbe means color, and zentrum center) is a type of crystallographic defect in which an anionic vacancy in a crystal is filled by one or more unpaired electrons.

See also: https://en.wikipedia.org/wiki/F-center

Crystal/lattice plane and direction notation

Although there is no strict consensus among the different research groups, we will use the terminology used by cristallographers.

(hkl): is used for a specific crystal face. e.g. (111) is the octahedral face intersecting the positive a, b and c axis

[hkl]: is used for a specific direction or plane normal. e.g. [100] denotes the positive a-axis direction

{ hkl }: is used for a family of equivalent crystal faces e.g. { 111 } denotes all faces of the octahedron in a cubic crystal

<hkl>: is used for a family of equivalent directions e.g. <111> denotes all directions normal to an octhedral face in a cubic system

See also: https://en.wikipedia.org/wiki/Miller_index

Some groups may use the small brackets (), <> for elements and the large brackets { },[] for the family of equivalent elements. The context of the phrase should make this clear.

Photochromic

A photochromic substance exhibits photochromism. The ability to change colour upon exposure to electromagnetic radiation.

See also: https://en.wikipedia.org/wiki/Photochromism

Gekoppelde termen in woordenlijst

F-centra, Spectrometers

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Hoofdstuk 2 - Group overview
Hoofdstuk 3 - Cont.
Hoofdstuk 5 - Hackmanite
Hoofdstuk 6 - Photochromic centra

Luminescence

Luminescence in this book is the spontaneous emission of light after stimulation by electromagnetic energy.

See also: https://en.wikipedia.org/wiki/Luminescence

Gekoppelde termen in woordenlijst

F-centra, Spectrometers, Vibronic

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Hoofdstuk 2 - Replacements

Hoofdstuk 5 - Hackmanite

Spectrometers

A spectrometer (/spɛkˈtrɒmɪtər/) is a scientific instrument used to separate and measure spectral components of a physical phenomenon. Spectrometer is a broad term often used to describe instruments that measure a continuous variable of a phenomenon where the spectral components are somehow mixed. In visible light a spectrometer can for instance separate white light and measure individual narrow bands of color, called a spectrum.

See also: https://en.wikipedia.org/wiki/Spectrometer

Gekoppelde termen in woordenlijst

EDX/SEM, F-centra, Luminescence, Photochromic, Raman spectroscopy, Vibronic

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Hoofdstuk 2 - Replacements

Ultramarine

Ultramarine is a deep blue color and a pigment which was originally made by grinding lapis lazuli into a powder.[1] The name comes from the Latin ultramarinus, literally "beyond the sea", because the pigment was imported into Europe from mines in Afghanistan by Italian traders during the 14th and 15th centuries.

See also: https://en.wikipedia.org/wiki/Ultramarine

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Hoofdstuk 3 - Applications

Syenite

Syenite is a coarse-grained intrusive igneous rock with a general composition similar to that of granite, but deficient in quartz, which, if present at all, occurs in relatively small concentrations (<5%). Some syenites contain larger proportions of mafic components and smaller amounts of felsic material than most granites; those are classed as being of intermediate composition. The volcanic equivalent of syenite is trachyte.

See also: https://en.wikipedia.org/wiki/Syenite

Gekoppelde termen in woordenlijst

Metamorphic, Pegmatite, Phonolitic, Plutonic rocks, Silicate

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Hoofdstuk 3 - Applications
Hoofdstuk 5 - Cont'd

Silicate

In chemistry, a silicate is any member of a family of anions consisting of silicon and oxygen. Silicates are extremely important materials, both natural (such as granite, gravel, and garnet) and artificial (such as Portland cement, ceramics, glass, and waterglass), for all sorts of technological and artistic activities.

See also:

https://en.wikipedia.org/wiki/Silicate

http://www.galleries.com/Silicates

http://www.science.uwaterloo.ca/~cchieh/cact/applychem/silicate.html

https://www.britannica.com/science/silicate-mineral

Gekoppelde termen in woordenlijst

Syenite, Zeolite

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Hoofdstuk 4 - Crystal structure
Hoofdstuk 5 - Chemistry and occurrence
Hoofdstuk 5 - Cont'd

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Gekoppelde termen in woordenlijst

Silicate

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Hoofdstuk 4 - Crystal structure

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Gekoppelde termen in woordenlijst

Dodecahedral, Lattice parameter, Plane and direction notation, Space group, Twinning

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Gekoppelde termen in woordenlijst

Dodecahedral, Point group, Space group, Twinning

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Hoofdstuk 4 - Lattice

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Gekoppelde termen in woordenlijst

Lattice parameter, Point group, Space group

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Hoofdstuk 4 - Crystal habit

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Gekoppelde termen in woordenlijst

Lattice parameter, Plane and direction notation, Point group, Space group

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Gekoppelde termen in woordenlijst

Metamorphic, Pegmatite, Phonolitic, Syenite

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Hoofdstuk 5 - Chemistry and occurrence

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Gekoppelde termen in woordenlijst

Metamorphic, Phonolitic, Plutonic rocks, Syenite

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Hoofdstuk 5 - Chemistry and occurrence

Hoofdstuk 5 - Cont'd

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Gekoppelde termen in woordenlijst

Pegmatite, Phonolitic, Plutonic rocks, Syenite

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Hoofdstuk 5 - Chemistry and occurrence
Hoofdstuk 5 - Cont'd
Hoofdstuk 7 - The Koksha minerals

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Gekoppelde termen in woordenlijst

Metamorphic, Pegmatite, Plutonic rocks, Syenite

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Hoofdstuk 5 - cont'd

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Gekoppelde termen in woordenlijst

Spectrometers, Vibronic

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Hoofdstuk 5 - Cont'd
Hoofdstuk 6 - Raman EDX

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See also: https://en.wikipedia.org/wiki/Vibronic_spectroscopy

Gekoppelde termen in woordenlijst

Luminescence, Raman spectroscopy, Spectrometers

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Hoofdstuk 6 - Cont'd

Hoofdstuk 6 - The cage problem

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See also:

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Gekoppelde termen in woordenlijst

Spectrometers

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Hoofdstuk 6 - Raman EDX

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Gekoppelde termen in woordenlijst

Luminescence, Photochromic, Spectrometers

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Hoofdstuk 6 - Photochromic centra

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Gekoppelde termen in woordenlijst

Point group, Space group, Twinning

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Hoofdstuk 4 - Crystal habit
Hoofdstuk 4 - Crystal habit