Blue, octahedral-substituted beryl from Norway: A case study of beryl and associated phenakite from Nedre Lapplægret, Drag i Tysfjord, Nordland

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Introduction

Beryl, ideally $Be_3Al_2Si_6O_{18}$, is the most wide-spread beryllium mineral. It occurs typically in pegmatites associated with granitic rocks. The dominant feature of its hexagonal structure is the rings of six corner-sharing SiO_4 tetrahedra, vertically stacked and forming hollow channels along the *c* axis. The hexagonal rings are connected laterally and vertically through Al in octahedral coordination and Be in a distorted tetrahedral environment. Some beryls may have appreciable amounts of Na and Cs, less commonly K and Rb, in the channels. This is associated with charge imbalance elsewhere in the structure, principally by substitution of Al by divalent elements at the octahedral sites and partial replacement of Be by Li at the tetrahedral sites. Some water is also commonly located in these channels. The so-called "octahedral" beryls or o-beryls show appreciable substitution of Fe^{2+}, Fe^{3+} and Mg for Al at the octahedral sites. The term o-beryl was introduced by Bakakin *et al*. (1967). A general formula of beryl, taking these substitutions into account, may be written $(\square,Na,Cs)(Be,Li)_3(Al,Fe^{3+},Fe^{2+},Mg)_2Si_6O_{18}\cdot nH_2O$. In o-type beryls, electron charge-transfer between di- and trivalent iron gives rise to a strong blue colour that is very characteristic (Fig. 1). The occurrence of this type of beryl in Norway is the subject of the present contribution. Wet-chemical analytical data, X-ray powder-diffraction pattern, density, refractive indices and infrared spectrum are presented for blue beryl associated with phenakite from the Nedre Lapplægret granitic pegmatite, Drag i Tysfjord, Nordland.

Blue beryl worldwide

A bluish beryl with unusually high contents of Fe, Mg and Na was first described from a pegmatite dike in Arizona (Schaller *et al*., 1962). It was also unusual with respect to the high Cs_2O content of 6.68 wt%. Several occurrences of such o-beryls were subsequently described, although generally not with high Cs_2O contents. A selection of some publications is included next, although reference to the Russian literature is omitted for space reasons. Fontan & Fransolet (1982) presented a wet-chemical analysis of blue beryl from the hematite mine of Lassur, Ariège, France. Aurisicchio *et al*. (1988) described dark blue beryl from the Calcaferro mine, Tuscany, Italy, and from a feldspatic vein in gneiss from the south side of Mount Cervandone, Piedmont, Italy. A deep azure-blue beryl was reported from quartz veins in conglomerate from southeast Ireland (Sanders & Doff, 1991). Blue beryl was found in Alpine veins in Tessin, Switzerland (Weiß, 1994). In quartz veins in syenite from the southern Yukon Territory, Canada, dark blue beryl and aquamarine occur (Groat *et al*., 2010, referring to an earlier report from 2003). An (Na,Mg)-enriched beryl is known from an NYF pegmatite in the Czech Republic (Novák & Filip, 2010). Přikryl *et al*. (2014) studied Fe-Mg-bearing beryl from granitic pegmatites in western Moravia, western Bohemia, Austria, Italy and Canada. This survey shows that blue o-beryls are by no means restricted to granitic pegmatites. It should be noted that the gemstone variety *aquamarine* with its greenish blue to pale blue colour has rather low contents of Fe and Na. It is therefore unfortunate that Groat *et al*. (2010) use the expression “dark blue aquamarine” in the title of their paper.

At this point it ought to be mentioned that there exists an Fe^{3+} analogue of beryl, stoppaniite, with end-member formula $Be_3Fe^{3+}{}_2Si_6O_{18}$ (Della Ventura *et al*., 2000). The mineral contains a fair amount of Fe^{2+}, as well as Mg, Al and Na, and the colour is blue. The Sc analogue of beryl is bazzite, nominally $Be_3Sc_2Si_6O_{18}$, which has a blue colour very similar to that of o-beryls, owing to minor Fe^{3+} and Fe^{2+} contents. However, both stoppaniite and bazzite have larger unit cells than o-beryl, and can be distinguished by X-ray diffractometry.

Fig. 1. Blue beryl from Badakshan, Afghanistan (bing.com/images).

Blue beryl and phenakite from Nedre Lapplægret

Sample description and preparation

The mineralogy of the granitic pegmatites within the Tysfjord granite, including the Nedre Lapplægret locality, was described by Husdal (2008). The specimens from Nedre Lapplægret used in the present investigation were collected by the author in 1968. Aggregates of short-prismatic, subhedral to euhedral phenakite crystals, up to 3 mm long, were surrounded by strongly blue beryl in irregular masses to 1 cm in size and as crystals up to 7 mm long. The beryl is green to yellowish green close to fluorite, but gives the same X-ray powder pattern with split lines as the blue variety (see below). Other minerals in the paragenesis are biotite, quartz, plagioclase and muscovite.

Five samples were crushed in a jaw crusher, then repeatedly in a disc grinder, and the material was sieved to 90 mesh. The dust fraction was removed by washing, and the sample was dried in a heating cabinet at low temperature. The minerals were separated into twelve fractions, numbered I to XII, using 1,1,2,2,-tetrabromoethane (density 2.967 g/cm^3), gradually diluted with acetone. Each of the twelve fractions was treated in a Franz isodynamic magnetic separator, varying the current in four to six steps. Biotite was removed in fraction I at 0.1 to 0.4 A. Fractions II to V consisted of phenakite; fraction II, magnetically separated at a current above 1.5 A, was chosen for further examination. Phenakite has a density slightly below that of the undiluted heavy liquid (see below). Some muscovite was separated in fraction VI at 0.35 to 1.0 A, together with the first fraction of beryl. More beryl followed in the next fractions, and fraction VIII at 0.9 to 1.2 A was selected for further study, constituting the largest amount of beryl obtained in the separation process. Finally, quartz and then plagioclase followed in the last fractions XI to XII.

X-ray powder data

Powder-diffraction data for beryl from Nedre Lapplægret, fraction VIII 0.9-12 A, are shown in Table 1. They were obtained using a Guinier–de Wolff quadruple focussing camera with 22.9 cm effective diameter, quartz monochromator and Fe*K*α$_1$ radiation (λ = 1.93728 Å). Lead nitrate was used as

Table 1. X-ray powder-diffraction data for blue beryl from Nedre Lapplægret and yellow beryl from Birch Portage (Radcliffe & Campbell, 1966).

Nedre Lapplægret (fraction VIII 0.9-1.2 A)				Birch Portage, Saskatchewan		
I_{obs}	d_{obs}(Å)	d_{calc}(Å)	*hkl*	I_{obs}	d_{obs}(Å)	*hkl*
70	7.97	8.01	100	100	7.94	100
30	4.59	4.59	002	31	4.567	002, 110
30	3.982	3.982	102	32	3.810	200, 102
100	3.256	3.257	112	62	3.251	112
5	3.029	3.026	210	19	3.011	202, 210
10	3.021	3.017	202			
100	2.874	2.874	211	50	2.8624	211
10	2.671	2.668	300	3	2.6611	300
20	2.525	2.526	212	16	2.5183	212
10	2.295	2.295	004	7	2.3007	004, 220
5	2.224	2.220	310	7	2.2097	310, 104
5	2.207	2.206	104			
10	2.159	2.158	311	11	2.1493	213, 311
<5	2.155	2.152	213			
5	1.999	1.999	312	3	2.0575	222, 114
10	1.990	1.991	204	16	1.9807	400, 204
<5	1.837	1.837	320	2	1.8305	320, $\underline{214}$
20	1.797	1.797	313	14	1.7937	321, 313
10	1.749	1.747	410	14	1.7378	410, 304
20	1.741	1.740	304			
10	1.718	1.716	411	9	1.7104	$\underline{322}$, 411
30	1.629	1.629	224	12	1.6261	224, 412
5	1.602	1.601	500	5	1.5946	500, 314
10	1.575	1.575	323	3	1.5730	215, $\underline{702}$
5	1.570	1.570	215			
10	1.531	1.530	006	2	1.5315	006, $\underline{330}$
30	1.519	1.517	413	8	1.5136	$\underline{404}$, 413

Underlined indices are too weak to be observed according to calculated data.

internal standard, and the intensities were visually estimated. Indexing of the reflections is in accord with calculated patterns (*e.g.* Borg & Smith, 1969). Refined cell parameters, using all 27 measured reflections, are a = 9.24(1) and c = 9.181(3) Å. The c:a ratio is 0.994, which is within the range of 0.991–0.996 typical for o-beryls (Aurisicchio *et al.*, 1988). For comparison, powder data for an ordinary yellow beryl from a Canadian locality (Radcliffe & Campbell, 1966) is included in Table 1 (obtained with Cu$K\alpha_1$ radiation). It has cell parameters a = 9.2091(6) and c = 9.1927(15) (Radcliffe, 1969), giving a c:a ratio of 0.998, within the narrow range of 0.997–0.998 given for "normal" beryls by Aurisicchio *et al*. (1988). The lower number of measured reflections for the yellow beryl compared to the blue beryl in Table 1 is striking. This is because the a and c parameters of ordinary beryl are nearly equal, and several reflections therefore overlap, hence the double sets of indices for the yellow beryl. The a parameter of beryl increases with incorporation of Fe and Mg for Al, and the individual reflections are resolved as closely spaced lines (we say that the lines in the powder pattern of blue beryl are "split"). This was first described by Bakakin *et al*. (1970). It is therefore easy to distinguish the two types of beryl by X-ray powder diffractometry. Considering calculated powder patterns for beryl (*e.g.* Borg & Smith, 1969), some of the reflections given for the yellow beryl in Table 1 (with indices 214, 322, 702, 330 and 404) are actually too weak to be observed ($I < 1$).

X-ray powder-diffraction data for phenakite (fraction II >1.5 A) were obtained in the same way as described for blue beryl. Refinement of cell parameters from 27 indexed reflections gave a = 12.468(3) and c = 8.252(2) Å.

Chemical composition

Wet-chemical analysis of blue beryl (fraction VIII 0.9-1.2 A) from Nedre Lapplæret was performed in 1976 by Brynjolf Bruun, then chemist at the former Mineralogical-Geological Museum. Analyses like this are not made any more, and the presentation of the result (Table 2) is the main reason for writing this article (it was intended to be published a long time ago). The elements Na, K and Li were determined by flame photometry, Mn, P and V by spectrophotometry and the remaining elements by gravimetrical methods (Be as phosphate). Water was determined as weight loss on ignition. H_2O^- in Table 2 means water lost below 105ºC; it could be mainly adsorbed and may be neglected (but consider the density determinations reported below); H_2O^+ is lost above this temperature and can be assumed to be hosted in the channels of the structure. Leif B. Garmann determined two important trace elements by neutron activation analysis: Sc 83 ppm, Cs 136 ppm.

The following nicely stoichiometric formula can be derived from the analytical data in Table 2, based on O = 18: $(Na_{0.322}K_{0.016})_{\Sigma 0.338}(Be_{2.994}Li_{0.038})_{\Sigma 3.032}(Al_{1.639}Fe^{2+}_{0.145}Mg_{0.139}Fe^{3+}_{0.100}Mn_{0.002}Ti_{0.002}Ca_{0.002})_{\Sigma 2.029}(Si_{5.957}P_{0.008})_{\Sigma 5.965}O_{18}\cdot 0.607H_2O$. The water content was calculated separately based on the H_2O^+ value. The total channel content, $\Sigma(Na+K+H_2O)$, is 0.945, close to unity.

Phenakite (fraction II >1.5 A) was not chemically analysed, since the mineral normally has a composition close to Be_2SiO_4. An optical spectrogram indicated minor amounts of Al, Fe, Mn, Mg and Ca.

Table 2. Chemical composition of blue beryl (fraction VIII 0.9-1.2 A) from Nedre Lapplægret, Drag i Tysfjord, Nordland.

Weight % oxide		Atoms on O = 18	
SiO_2	63.19	Si	5.957
BeO	13.22	Be	2.994
TiO_2	0.03	Ti	0.002
Al_2O_3	14.75	Al	1.639
Fe_2O_3	1.40	Fe^{3+}	0.100
FeO	1.84	Fe^{2+}	0.145
MnO	0.02	Mn	0.002
P_2O_5	0.02	P	0.008
V_2O_5	<0.05		
MgO	0.99	Mg	0.139
CaO	0.02	Ca	0.002
Na_2O	1.76	Na	0.322
K_2O	0.13	K	0.016
Li_2O	0.10	Li	0.038
H_2O^-	0.48		
H_2O^+	1.93		
Total	99.88		

Analyst: B. Bruun 1976 (Mineralogisk-Geologisk Museum, Oslo).
Sc 83 ppm, Cs 136 ppm (analyst: L.B. Garmann).

Optical data and density

Refractive indices were measured in white light, and the refractive index of the immersion liquids was checked with an Abbe refractometer. Blue beryl (fraction VIII 0.9-1.2 A) is uniaxial negative with $\omega = 1.591 \pm 0.001$ and $\varepsilon = 1.586 \pm 0.001$. The mineral is weakly pleochroic with ω nearly colourless and ε faintly blue. Refractive indices for phenakite (fraction II >1.5 A) are $\omega = 1.653 \pm 0.001$ and $\varepsilon = 1.667 \pm 0.001$; it is uniaxial positive.

Density was measured on separated material with a 25 ml pycnometer, using toluene. For blue beryl (sample weight 14.3453 g), $d = 2.767$ g/cm^3, and for phenakite (15.3195 g), $d = 2.962$ g/cm^3. Density calculated for phenakite, assuming the ideal composition Be_2SiO_4, is 2.962 g/cm^3. Density calculated from the empirical formula of blue beryl is 2.756 g/cm^3. However, if H_2O^- is taken into account as a channel constituent, d_{calc} is 2.769 g/cm^3, in better agreement with the measured value. This could possibly indicate that some of the water lost at fairly low temperatures may actually reside in the channels. A DTA run in air with Al_2O_3 as reference material was performed on the blue beryl with a Du Pont instrument, up to 1170ºC. Two distinct endothermal peaks occur at 115 and 175ºC, the former being larger than the latter. The X-ray powder pattern was unchanged after heating.

The empirical Gladstone-Dale relationship can be applied to test the consistency between chemical composition, mean refractive index and measured density of a mineral. To this end, the compatibility index $CI = 1 – K_p/K_c$ is used, where K_c is computed from chemical analytical data and K_p from refractivity and density. Ideally, K_p should equal K_c. Data presented here for blue beryl from Nedre Lapplægret give CI = 0.009 (including H_2O^+ in the computation of K_c), which is rated as superior by Mandarino (1981). For phenakite, CI = 0.002, also a superior value. The revised *k* parameters of Mandarino (1981) were used in the calculation of K_c.

Co-existing beryl and phenakite

The association of beryl and phenakite has been reported in a number of cases, but the interpretation of their mutual relationship differs. Phenakite being generated as a late-stage phase by alteration of beryl has been documented in some cases (*e.g.* Jonsson & Langhof, 1997). On the other hand, beryl was supposed to have formed from phenakite by "addition of SiO_2" in the aplitic border of a granite pluton during a late magmatic phase of emplacement (Abrecht & Hänni, 1979). However, the presence of Al would seem to be more critical in this respect. The beryl has a composition typical of o-beryls. Husdal (2008) reported beryl associated with phenakite from the Nedre Lapplægret and Håkonhals pegmatites and suggested that at least some of the beryl from the latter locality formed during breakdown of phenakite in a deformation process. As for the specimens from Nedre Lapplægret examined by the present author, the two minerals appear to co-exist without any signs of replacement. Phenakite seems to have crystallized prior to beryl. Generally, it is the availability of Al that determines if phenakite or beryl will form.

Blue o-beryl in Norway

As mentioned above, o-beryls can conveniently be distinguished from ordinary beryl by the splitting of certain lines in the X-ray powder-diffraction pattern. The archive of X-ray powder photographs of beryl in the former Mineralogical-Geological Museum, obtained with 9 cm Debye–Scherrer cameras, has been searched. Patterns of Norwegian beryls with split reflections and their localities are listed below (with exposure or "film" numbers and localities as written in Norwegian on the "film" envelopes):

1110 – Svartisen
15931 – SE Kleppsvann, Tørdal, Telemark (slightly enlarged cell, possibly some Sc, but not bazzite)
15981 – Bjørnefoss, Flatisvann, Svartisen
15982 – Under Svartisen opp for Langevatn
16055 – Kleppen, Tørdal, Telemark
16303 – Søftestad, Nissedal, Telemark
16368 – Fra pegmatittgang, Egersundstrakten
16393 – Stetind feltspatforekomst (only slight splitting of reflections)
18974, 18975, 23692 – Nedre Lapplægret, Drag i Tysfjord

It is remarkable that pegmatitic, blue o-beryls are mostly confined to two areas in northern Norway, Tysfjord and Svartisen, and are not known from the major areas of beryl-bearing pegmatites in southern Norway, like Iveland–Evje and Østfold. The pegmatites in the Tørdal area are an exception, as both o-beryl and bazzite are known from there. The reason for the restricted occurrence of o-beryl is not obvious, but could be related to assimilation of mafic bedrock rich in Fe and Mg. Blue beryl from the Svartisen pegmatites may reach considerable sizes; long-prismatic crystals several dm in length and stubby crystals to 10 cm in diameter are known. Unfortunately, the exact locality of the pegmatite near Egersund is not known (the specimen was acquired by the museum from a Mr Tybring in 1895). At the Søftestad iron mine, rather pale blue o-beryl occurs along fissures in the bedrock.

Infrared spectra of some Norwegian beryls

Water in the channels of the beryl structure can occur in two different orientations, depending on the presence or absence of alkali ions (Fig. 2). The water molecule has a weak dipole character, which means that the charge is displaced so that the oxygen end of the molecule is slightly negative, and the hydrogen ends are correspondingly positive. In the absence of alkali ions, the water molecule is oriented with its diad axis perpendicular to the *c* axis of the beryl, and the hydrogen atoms point towards the oxygen atoms of the channel wall. This is called type I water (Fig. 2). In the presence of alkali ions, the water molecule is oriented with its negative end pointing towards the positive alkali ion, and its diad axis is parallel to the *c* axis of the crystal. This is type II water (Fig. 2). Of course, both orientations may occur in a given crystal, depending on the ratio between water molecules and alkali ions.

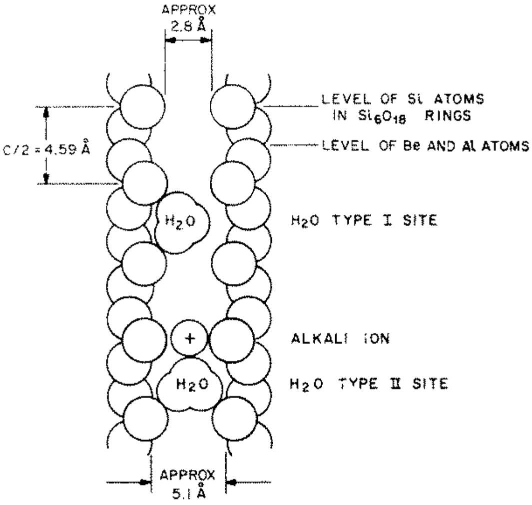

Fig. 2. Side view of the channel in the beryl structure (the *c* axis is vertical), showing the two types of water molecule orientation in relation to the surrounding oxygen atoms and the presence of an alkali ion. After Wood & Nassau (1967).

In infrared absorption spectra of beryl, water molecules of type I and II produce stretching and bending vibrations at different frequencies (Wood & Nassau, 1967; Aurisicchio *et al.*, 1994). A selection of infrared spectra of Norwegian beryls is presented in Fig 3. They were recorded in the 4000 to 625 cm^{-1} region using a Perkin-Elmer spectrometer. Finely ground beryl samples were dispersed in Nujol oil on NaCl discs, and roughly equal amounts of material were used for each recording. The two missing regions of the curves in Fig. 3 are where the Nujol oil absorbs infrared radiation. The Nedre Lapplægret beryl is shown as spectrum **e** at the bottom of the figure.

The O–H stretching mode of water I occurs at 3700 cm^{-1}, of water II at 3600 and 3660 cm^{-1}. The spectra in Fig. 3 are arranged with increasing content of water II and decreasing content of water I from top to bottom, which must coincide with an increasing amount of alkali ions. It can also be seen that the bending vibration of water II at 1635 cm^{-1} is increasing concurrently. The bending vibration of water I at 1600 cm^{-1} shows very little variation in size. The slight inflection of the curves at around 2350 cm^{-1} is probably due to a minor content of CO_2 in the channels (Wood & Nassau, 1967). In conclusion, infrared spectra are good indicators of the amount of alkali ions in the beryl structure.

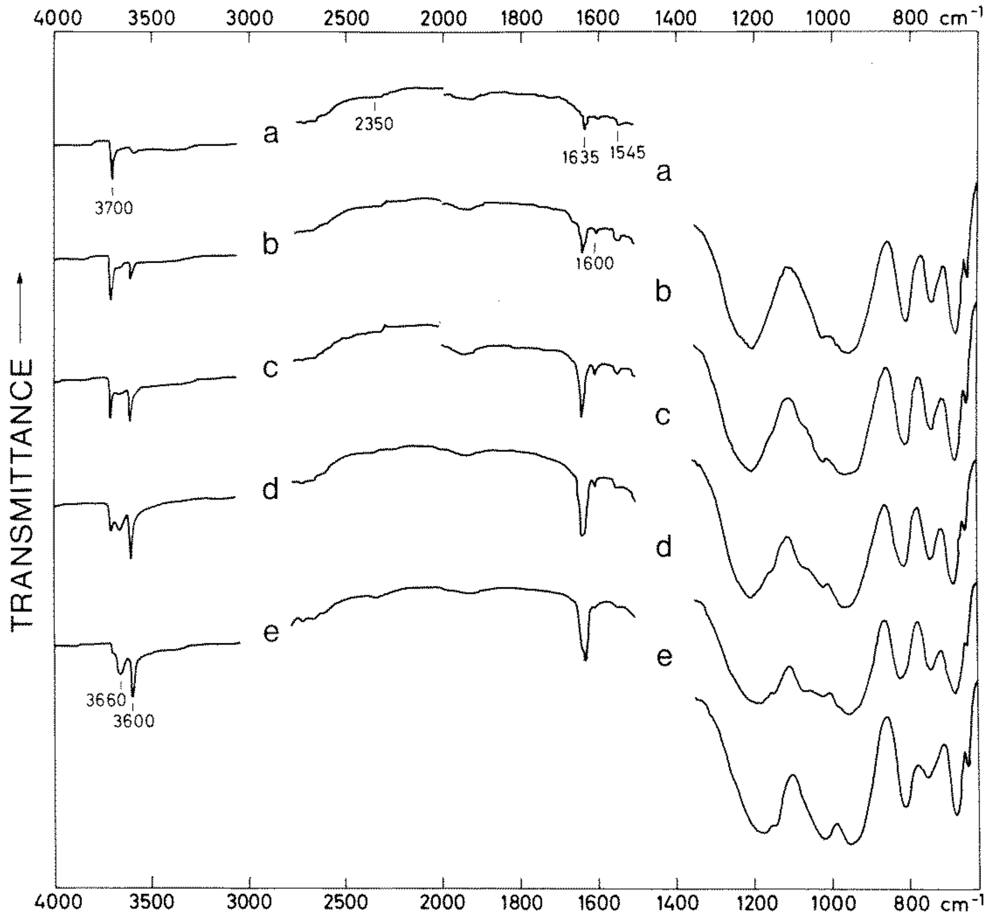

Fig. 3. Infrared absorption spectra of Norwegian beryls. (a) Green beryl from Unneland, Evje, Aust-Agder. (b) Yellow, transparent beryl from Høydalen, Tørdal, Telemark. (c) Rose, transparent beryl from Høydalen, Tørdal, Telemark. (d) Rose, transparent beryl from Ågskaret, Holandsfjord, Nordland. (e) Blue beryl (fraction VIII 0.9-1.2 A) from Nedre Lapplægret, Drag i Tysfjord, Nordland. The spectrum of bluish beryl from Håkonhals, Hamarøy, Nordland, is similar. The content of alkalis situated in the channels increases from (a) to (e) (see text).

Acknowledgements

Sveinung Bergstøl worked on beryls with split lines in their X-ray powder patterns in 1965-1966, by X-raying a number of blue beryls from the collections of the Mineralogical-Geological Museum and searching the archive of Debye-Scherrer powder photographs at the museum. He entrusted the present author to continue this work. Brynjolf Bruun and Leif B. Garmann made major contributions by supplying chemical analytical data. I am greatly indebted to Roy Kristiansen who recorded a number of infrared spectra of Norwegian beryls, of which five are included here.

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