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On the occurrence of gallium and germanium in the Bergslagen ore province, Sweden

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ABSTRACT

The presence of the critical and sought-after (semi-)metals gallium (Ga) and germanium (Ge) has previously been reported from mineralisations in the Bergslagen ore province, south central Sweden. Some of these reports were however recently shown to be questionable or erroneous. Here we summarise early analytical work on these metals in mineral deposits of the Bergslagen province, as well as briefly report new analytical data for Ga and Ge from recent, in part on-going work on different mineralisation types. The new data show that the sampled sulphide and iron oxide mineralisations in the Bergslagen province are overall not particularly enriched in Ga, and even less so with regards to Ge. One major exception is the significant Ga enrichment observed in skarn-hosted Fe-REE-polymetallic deposits of Bastnäs type. Notably, these mineralisations also host increased contents of Ge. Based on this broader suite of sampled deposits, the suggested correlation between Ga and Al contents in previously studied material with relatively increased Ga grades, is in part contradicted, indicating that Ga is only in part sequestered through straightforward Al-substitution into aluminium silicate and oxide minerals. The mineralisations that do exhibit significantly increased Ge contents, in addition to the Bastnäs-type deposits, are represented by both sulphide-dominated ones and Fe (-Mn) oxide-rich systems.

INTRODUCTION AND BACKGROUND

The (semi-)metals gallium (Ga) and germanium (Ge) are both extensively used in a wide range of high-technology and energy-saving applications, such as in the production of semi-conductors including integrated circuits, photovoltaics, as well as modern LEDs and related energy-saving products (e.g., Gunn 2014). As intra-European mine production of these metals is essentially lacking and their demand is only satisfied through imports that are fraught with issues of risk, they are presently deemed as critical to the European industry, and hence included in the current list of most critical raw materials (EU commission 2017).

The presence of Ga and Ge in the ores of the Zinkgruvan mine in the southernmost part of the Palaeoproterozoic Bergslagen ore province, Sweden, was recently brought up to discussion by Jansson et al. (2016). By means of unpublished and new analytical datasets, they questioned the previously stated significant enrichment of Ga and Ge in this deposit (SGU 2003; Jonsson et al. 2015). Here, we review the historical analytical data available for these metals in Bergslagen, and add new data regarding their occurrence in a suite of mineralisations of contrasting types.

Gallium and germanium in the mineral deposits of Bergslagen

Gallium was originally discovered in sphalerite from a sulphide deposit in France and described by Lecoq de Boisbaudran in 1875; very soon thereafter, in 1876, he published a further study on sphalerites from a suite of localities, including a sample from the Zinkgruvan mine in the southernmost part of the Bergslagen ore province (Fig. 1). This he reported to be Ga-bearing, albeit poorly so; “très pauvres” (Lecoq de Boisbaudran 1876). It seems not unlikely that this early mention may to some extent have contributed to the “reputation” of Ga in the Zinkgruvan sphalerite ore.

Much later, Papish & Stilson (1930), utilising graphite and silver arc spectrography, determined the presence of Ga in gahnite (zinc spinel, ideally ZnAl₂O₄) from the Falu mine, northern Bergslagen (as well as in gahnite from Orijärvi, Finland). During a wide-ranging study of different minerals, the presence of Ga in Falu mine gahnite was further corroborated by Goldschmidt & Peters (1931), who also identified it as present in braunite (ideally Mn²⁺Mn³⁺SiO₄) from the Åker marble quarry in westernmost Bergslagen, as well as in sensu stricto spinel (ideally MgAl₂O₄) from the Åker marble quarry in the south-eastern part of the province. A little later, Landergren (1935), by means of X-ray spectrophotometry, determined Ga contents of about 30 ppm in bulk ore from the Saxberget mine, and less than 1 ppm in ore from the Garpenberg mine. He also, quite succinctly, expressed his belief that Ga at Saxberget was predominantly hosted by silicate minerals, rather than in the sulphidic part of the ore.
In a study by Stoiber (1940) neither Ga nor Ge were detected in his new analyses of Zinkgruvan ("Ammenberg") material, but interestingly, a sample from the Dannemora mine “South section” yielded a coarse determination of its Ge content, given as <0.01 %, i.e. positively identified, at less than 100 ppm.

A slightly later arc spectroscopy study by Gabrielson (1945) was focused on the presence of trace metals including Ga and Ge in sphalerite, and represents quite a wide range of occurrences and deposit types. This work was in part supported also by wet chemical analysis, for correlation purposes, and likely represented the first study approaching quantitative
analysis of several trace elements in Swedish sphalerites. Albeit with reservation for the low precision of the method applied, Gabrielson reported measured, anomalous Ga contents in sphalerites from some 25 (in part) sulphide-mineralised assemblages, and additional from different (Phanerozoic) sedimentary rocks; of these mineralisations, Doverstorp, Kaveltorp, Saxberget, Meltorp, Dannemora (all in Bergslagen), Bolditen, Bjurlden, Östra Hökull and Ravliden-Ravldmyran (the latter four from the Skellefte district) stand out as markedly increased in Ga, with stated contents at or above c. 10 ppm. Overall, 34 localities with measured Ga contents were reported by Gabrielson. Notably, in this study, analyses of Zinkgruvan (“Åmmebergsfältet”) sphalerite yielded only sparse contents at c. 4 ppm Ga. As referred to by Jansson et al. (2016), Henriques (1964) presented trace element analyses, including Ga (up to 10 ppm in Zn ore) and Ge (up to 100 ppm in Zn ore), performed by spectrography of ores and host rocks at Zinkgruvan.

Germanium, in turn, was originally discovered in 1886 as a major constituent of the silver-rich mineral argyrodite (ideally Ag₆Ge₆S₁₇) from Freiberg, Germany, and was later determined by several workers as a minor component in a number of sulphide minerals, and particularly in sphalerite, from different deposits (Doelter & Leitmeier 1926; Papish 1928, and references therein).

While Hadding (1922) had verified the presence of Ge in cassiterite from the Finno granitic pegmatite quarry, near Falun, Papish (1929) determined Ge in topaz from the nearby Broddbo pegmatite (boulder). The latter author also found Ge present in other minerals, from the fractionated pegmatite dykes at Utō, in the eastern part of Bergslagen; in “lepidolite” (Li-mica), spodumene and red, elbaitic tourmaline (“rubellite”).

In the study by Gabrielson (1945), referred to above, measurable Ge contents were reported in sphalerites from five discrete mineralisations: Doverstorp, Södra Fältet (Dannemora), Vattholmagruvan (Dannemora), Värmlands Taberg (all in Bergslagen) and Östra Hökull in the Skellefte district, in addition to two sphalerites from essentially non-mineralised Phanerozoic rocks. Of the former, the highest contents at (a stated) c. 100 ppm are represented by the two occurrences in the Dannemora field and Östra Hökull (one sample out of six analysed).

**New data and results**

Recently, we have sampled and analysed different mineralisation types in Bergslagen, both in the field (mainly from mine dumps) and from historical sample suites, with the aim to increase the knowledge on the distribution of rare and critical metals, including Ga and Ge.

Of the total set, 222 samples have been analysed for both Ga and Ge. The majority of the samples are mineralised, with some additional, mainly non-mineralised, altered host-rocks. The samples were analysed on different occasions over a few years time at the ALS laboratory in Vancouver, Canada. The major elements were analysed by ICP-AES following fusion, and most trace elements including Ga were analysed by ICP-MS after lithium borate fusion and nitric acid dissolution. Remaining trace elements, including germanium, were extracted by *aqua regia* prior to ICP-MS analyses. Due to the small size of some of the historical samples this preparation and analytical method were applied for all elements.

The recommended values for average upper crust concentrations of Ga and Ge are 17.5 and 1.4 ppm, respectively (Rudnick & Gao 2014). However, as can be seen in various datasets including our own, gallium concentrations of up to 25 ppm are not uncommon in the Palaeoproterozoic bedrock of the Bergslagen province, also at considerable distances from any known mineralisations. Therefore, only Ga and Ge concentrations double and higher than the recommended values for the average upper crust are reported here as Table 1. For simplicity of representation, these are furthermore classified into three broad categories of 2 to 10, >10 to 50 and >50 times these values, for either or both of Ga and Ge (Fig. 1). In the following text, these are referred to as “modestly increased”, “increased” and “markedly increased”. These are arbitrary classifications chosen for ease of applicability in a region with – so far – sparse information available on their endowment in mineralised systems.

Relative to the latter, the new data show several examples of increased gallium contents, which are essentially found within one particular type of deposit. During sampling and analyses of the REE-enriched Fe oxide (polymetallic) skarn mineralisations of Bastnäs type (Geijer 1961), which outline the so-called REE-line of the Bergslagen province (Jonsson & Högdahl 2013), a suite of samples was observed to contain significant gallium contents, as was also reported briefly by Högdahl et al. (2015). These have now been added to, and we find that the overall dataset from the Bastnäs-type skarns yield Ga contents from less than 35 ppm to up to nearly 1000 ppm (Table 1). The deposits located around the eponymous Bastnäs mine fields near Riddarhyttan in the central part of the REE line, have been reported here collectively as the “Bastnäs fields”, represent sampled material of REE-Fe-mineralised skarn and altered immediate host rocks from the Nya Bastnäs and Bastnäs Storgruva mines, while separating the partly “ore-quartzite”-hosted Fe oxide-sulphide mineralisation southwest thereof at Myrbacksfältet, as well as a mainly Cu sulphide-mineralised sample from Knuts Stoll. In the Norberg area, the Södra Hackspik, Östanmossa and Johanna mines, as well as the Malmkärre mine southwest of that town were sampled, and all yielded increased Ga contents, most markedly so in the cases of the Johanna (average 193 ppm) and Malmkärre (average 300 ppm) mines. In addition to these, samples from the REE-Fe oxide skarn at Rödbergsgruvorna, and the felsic metavolcanics (skarn)-hosted REE-Fe oxide mineralisation at Östra Gyttorpsgruvan, both west of Nora, were also included; of these, the latter yielded a Ga content at around 200 ppm.

In addition to the REE-line, increased to markedly increased Ga contents were also encountered in the polymetallic sulphide deposit at Skyttgruvan W of Falun (106 ppm), and in Fe-oxide mineralisations at Bispberg, near Säter (48 ppm), Klacka-Lerberg NW of Nora (35 ppm), as well as Skvatterberg (56 ppm) and Kopslabyttan S of Borlänge (average 40 ppm), the latter being an apatite iron oxide mineralisation (Geijer &
Table 1. New data on mineralisations in Bergslagen with Ga and Ge concentrations twice or more than that of the average upper crust.

| Mineralisation type | Locality                  | Ga\(^a\) (ppm) | Average | No. samples\(^c\) | Ge\(^b\) (ppm) | Average | No. samples\(^d\) |
|---------------------|---------------------------|----------------|---------|------------------|----------------|---------|------------------|
| Skarn Fe-REE        | Bastnäs fields            | 37–285         | 155     | 6(12)            | 3              | 1(12)   |
| Skarn Fe-REE        | Johanna                   | 98–308         | 193     | 3(3)             | 24–50          | 37      | 2(3)             |
| Skarn Fe-REE        | Malmkärра                 | 120–922        | 300     | 7(7)             | 3.8–131        | 31      | 6(7)             |
| Skarn Fe-REE        | Rödberg                   | 123            | 1(5)    | 2.9–7.8          | 5.3            | 2(5)    |
| Skarn Fe-REE        | Södra Hackspik            | 264            | 1(1)    | 17               | 1(1)           |
| Skarn Fe-REE        | Östanmossa                | 88–243         | 162     | 3(4)             | 6.8–15         | 11      | 3(4)             |
| Skarn Fe-REE        | Östra Gyttorp             | 202            | 1(1)    | 20               | 1(1)           |
| Cu-polyhalite       | Knutstjöll                | 56             | 1(1)    | <2*AUC           | 1(1)           |
| Cu-Co-Bi-Fe-REE     | Myrback field             | 50             | 1(5)    | 3.7              | 1(5)           |
| Apatite Fe-oxide    | Kopsjärvettan             | 37–43          | 40      | 2(3)             | <2*AUC         | 3(3)    |
| Fe-oxide            | Bispegården              | 48             | 1(1)    | <2*AUC           | 1(1)           |
| Fe-oxide            | Klacka-Lerberg            | 35             | 1(3)    | <2*AUC           | 3(3)           |
| Fe-oxide            | Skvatterberg              | 56             | 1(1)    | <2*AUC           | 1(1)           |
| Fe-oxide (BIF)      | Forsbo                    | <2*AUC         | 3(3)    | 3.0              | 1(3)           |
| Fe-oxide Cu-polyhalite | Sandbacken              | <2*AUC         | 1(1)    | 3.6              | 1(1)           |
| Cu-Fe polyhalite    | Ingelsgruvorna            | <2*AUC         | 1(1)    | 4.1              | 1(1)           |
| Cu-polyhalite       | Ö Hårddalaren            | <2*AUC         | 2(2)    | 4.2              | 1(2)           |
| Cu-polyhalite       | Stripásen                | <2*AUC         | 5(5)    | 4.1–5.8          | 4.9            | 3(5)    |
| Pb-Zn polyhalite    | Bronäs                    | <2*AUC         | 1(1)    | 3.7              | 1(1)           |
| Pb-Zn polyhalite    | Lahäll                    | 36             | 1(7)    | <2*AUC           | 7(7)           |
| Zn-polyhalite       | Skyttgruvan               | 106            | 80      | 2(2)             | 5.8            | 1(2)    |

\(^a\)Average reference values of Ga and Ge concentrations for the upper crust are 17.5 and 1.4 ppm, respectively (Rudnick & Gao 2014).

\(^b\)Range of Ga concentrations twice or more than the average upper crust.

\(^c\)Number of samples with Ga concentrations twice or more than the average upper crust. Total number of analysed samples in brackets.

\(^d\)Range of Ge concentrations twice or more than the average upper crust. Total number of analysed samples in brackets.

\(<2*AUC = Less than twice the concentration of Ga and Ge, 17.5 and 1.4 ppm respectively, than the average upper crust as given by Rudnick & Gao (2014).

Magnusson 1944; Jonsson et al. 2015). The granitic pegmatites included among the analysed samples also show increased to markedly increased Ga contents (Holmtjärn at 44 ppm, and Vimmelmora at 53 ppm, respectively).

In the case of Ge, the new dataset shows increases relative to the upper continental crust in a number of different deposits, mostly represented by Bastnäs-type skarn deposits, with averages at around 5–37 ppm, but locally up to 131 ppm, represented by one of the samples from Malmkärра.

Only a few of the diverse sulphide mineralisations that were analysed showed, with one exception, at best modest increases in Ge. These include the carbonate-hosted sulphides at Bronäsgruvan (Sala), the skarn-hosted Stripåsen and Sandbacken mines (Norberg), the banded iron formation (BIF) deposit at Forsbo (Säter area) as well as the Fe oxide and sulphide-bearing systems at Myrbacksfältet (Riddarhyttan) and Ingelsgruvorna (NE Guldmedshyttan) (Table 1, Fig. 1).

The two mineralogically very contrasting samples from the Falun area (Östra Hårdmalmerna, the quartz-dominated, Pb-Bi-Cu-Se sulphosalts-bearing and Au-enriched part of the Falu copper mine, and the Zn-mineralised skarn of Skyttgruvan-Näverberg, W of Falun proper, noted for its content of gahnite) exhibit increased and modestly increased enrichment in Ge, at 42 and 3.8 ppm, respectively.

Discussion and conclusions

The so far observed overall low concentrations of Ga combined with the apparent scarcity of significantly increased contents of Ge in the sulphidic mineral deposits of Bergslagen suggest that the available metal sources, or source regions, and processes responsible for mineralisation may not have been of a nature conducive to concentration of these elements. In part, this is to be expected in the case of sulphide-dominated systems, as characteristically both elements, and in particular Ge, tend to be more concentrated in low temperature deposits such as those of Mississippi Valley type (Bonnet et al. 2016, and references therein), as was also suggested early on by e.g. Stoibler (1940).

The observed reasonable correlation between Ga and Al (i.e., Al\(_2\)O\(_3\)), and contrastingly poor correlation between Ga and Zn in the Zinkgruvan samples presented by Jansson et al. (2016) suggest that the main sinks for Ga are Al-rich minerals rather than phyllosilicates. Prior to that, Jansson et al. (2013) had also found a similar correlation between Ga and Al in samples from the Stollberg field, NE of Ludvika, and attributed it to a detrital rather than hydrothermal input. Gallium incorporation into Al-bearing minerals, in which it can be substituted for Al\(^{3+}\), are likely to be dominated by silicates (e.g. Brandt & Kydd 1998; Mangir Murshed & Gesing 2008; Dittrich et al. 2011), yet also oxidic minerals including spinel (sensu lato) may be locally important hosts, as was shown by, e.g. Papish & Stilson (1930) and Goldschmidt & Peters (1931). In extreme cases, silicates such as chlorite (sensu lato) has been shown to carry over 20 wt% Ga\(_2\)O\(_3\) (Johan et al. 1983). A Ga- Al\(_2\)O\(_3\) plot (Fig. 2) utilising our full new dataset (222 samples) does not show any clear-cut correlation, thus suggesting that no singular substitution and/or host mineral can account for the observed Ga contents. Moreover, it must be stated that in order to positively prove such relations, chemical analyses of discrete minerals must be performed, rather than of bulk ore or rock samples. Bulk analyses will never fully show, or prove, specific mineral-related substitution mechanisms, as different potential host minerals (e.g. sulphides versus oxides versus silicates; all three being possible host structures for Ga) may be present. Notably, detailed insights as to the sequestration of Ga in the markedly
enriched mineralisations in the REE-line, such as Malmkärna, are still wanting.

Based on the diversity of mineralisations observed to exhibit overall increased Ge contents, we find it likely that, depending on deposit type, and their mineralogy as well as inherent process of formation, major host minerals for Ge may be equally variable (cf. Höll et al. 2007). It should be remarked in this context that the highest observed Ge contents among our samples were found in Cu sulphide-rich as well as in Fe-dominated oxide mineralisations. The exceptions found in the case of the observed, increased to markedly increased Ga and Ge contents in the skarn-hosted REE-Fe mineralisations of the REE line bear evidence that some remains potential for processes having concentrated these elements in the Bergslagen province. This also suggests that further study of particular (suites of) deposit types is warranted.

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