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Natürlicher Graphit: Vorkommen, Abbau und Aufbereitung

Natural graphite: occurrence, mining and processing

Bachelorarbeit

Bachelor's Thesis

von / presented by

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I thank my friends, my fellow members of the student board and my grandmother Mrs. Margarete Fahl for the steady encouragement and motivation. I am proud to present my final research results.

Abstract

This thesis is a global and general review of the mineral resource natural graphite. Natural graphite is a critical raw material with a high economic importance for the European union. Natural graphite is used in high technology applications like anodes for electric vehicle lithium-ion batteries. Main aim of the thesis is to provide an advanced understanding of the mineral resource natural graphite. Different scientific models and theoretical approaches of the natural graphite formation are presented. A coherent description and evaluation of the mineralogy, regional and local geology of occurrences and deposits in the European Union is given. The technical extraction and beneficiation process is outlined. Facts and linkages are visualized with flow schemes, tables and schematic graphics. Detailed case study examples illustrate and underline the topics of the thesis.

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1 Introduction

Intention and Motivation:

Our modern life style is based on the consumption of mineral resources. From cradle to grave we are surrounded by consumer products created out of stones and rocks. More and more elements from Pauling's periodic system are installed in our cars, devices or infrastructure**.** A growing world population, the economic growth in developing and emerging countries and a constant seek for innovation further enhances the demand for mineral resources. To satisfy this increasing demand our society needs to solve serval severe global challenges. Managing the worldwide mineral resources trade flow is one challenge. Establishing an attitude of awareness and responsibility for a sustainable development in the global mining and mineral resources industry is another challenge.

Natural graphite is defined as a critical raw material by the European union. Securing an independent European natural graphite supply apart from international trade networks for the European manufacturing industry is significant for the development of the European and German high technology economy. Natural graphite is mainly utilized for steel manufacturing but also in high technology applications like anodes for electric vehicle lithium-ion batteries. The growing electric vehicle production promotes the international interest in information about the mineral resource natural graphite.

The description and evaluation to the mineralogy, regional and local geology of occurrences and deposits in the European union in a coherent manner provides an important and advanced understanding of the mineral resource natural graphite. Giving an overview of the extraction methods and technical beneficiation process shall help to gain a comprehensive understanding of the mineral resource natural graphite and the production process.

Methodology:

Designed for a wide audience with diverse backgrounds in earth science, mining or mineral resource economics the thesis provides a general and global review of the mineral resource natural graphite. A short data sheet of the mineral resource natural graphite is designed to present the main features and information. Different scientific models and theoretical approaches of the natural graphite formation are presented and compared. A synopsis of the location of European occurrences and deposits is given. The mineralogy, regional and local geology and ore body layout of diverse occurrences and deposit types is outlined. Facts and linkages are visualized and graphically evaluated with flow schemes, tables, graphs and schematic graphics. A short glossary of terms and additional information charts are contained in the appendix. The technical process steps of natural graphite extraction and beneficiation is described by giving a detailed case study example and listing utilized aggregates. Satellite photographs of a North Korean mine are analyzed to vividly illustrate the environmental impact of natural graphite mining. Price charts are designed and analyzed to depict the volatile natural graphite market. A brief evaluation of the current criticality and an outlook on the future development of the natural graphite market is given. The thesis is concluded by an outlook on additional research potential.

2 Ressource: natural graphite

Table 2.1 explains the mineral features, the categorization, the worldwide mine production of 2014 and the estimated resources and reserves of natural graphite.

Table 2.1 Short description natural graphite. Data from (Indian Bureau of Mines, 2016, p. 44; USGS a, 2017, p. 75; Okrusch et al., 2010, p. 51; BGR, 2014, p. 2)

Mineral features

Element: C

Crystal structure: hexagonal (*6/m 2/m 2/m*) graphite 2 H or trigonal $\overline{3}$ m

Special features: high anisotropy, high conductivity, high intercalation strength

Herfindahl-Hirschman Index*: 5.015 (high)

Categorization

Amorphous - Carbon content 70 - 85 %

Crystalline flake - Carbon content 85 - 90 %

Vein-type - Carbon content 90 - 95 %

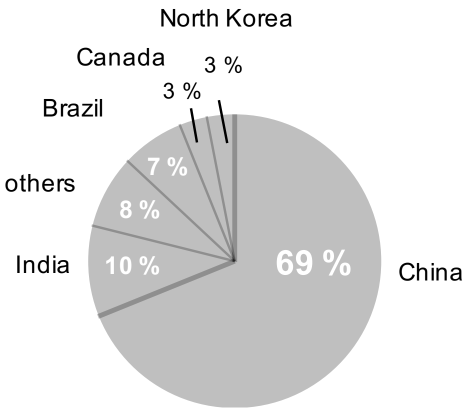
Worldwide mine production 2014

China 780,000 tonnes

India 170,000 tonnes

Brazil 80,000 tonnes

Overall 1.13 million tonnes*



Worldwide resources 2014: 800 million tonnes

Worldwide reserves 2014: 76.8 million tonnes

Natural graphite is an allotrope of pure carbon that crystallizes in the hexagonal crystal lattice, with rhombohedral symmetry. The six-sided, tabular crystals commonly have striated faces. Natural graphite is an excellent thermal and electric conductor. Natural graphite has a hardness of 1 to 2 according to the Mohs Scale of mineral hardness. The touch of natural graphite is soft and it has a greasy texture. Natural graphite has high intercalation strength. It will form intercalation compounds with alkali metal vapors. The metal ions, mostly potassium, rubidium, cesium and lithium fit between the planar carbon structure of the crystals. Natural graphite is defined as a critical raw material by the European Union and has a high HHI-Index of 5.015. (Taylor, 2006, p. 508)

Natural graphite is commonly categorized by particle size and chemical properties. Figure 2.1 explains the different types of natural graphite products and their size distribution. Figure 2.2 outlines the global natural graphite demand by application in 2012.

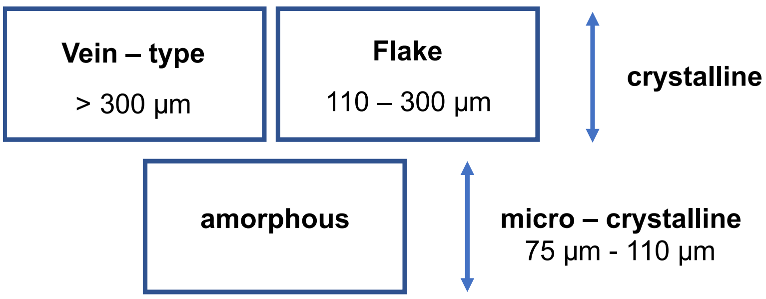


Figure 2.1 Overview: natural graphite product types. Data from (Scogings b, 2015, p. 79)

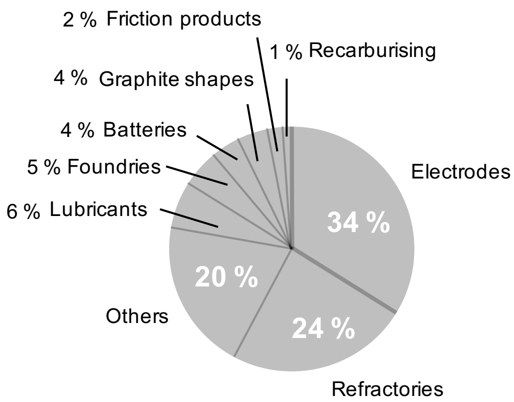


Figure 2.2 Global natural graphite demand by application in 2012. Modified from (Shaw, 2015, p. 6)

[*Appendix A: defines the HHI-Index; Appendix E: defines Units]

3 Occurrences - Deposits: of natural graphite

Mineral occurrences are geologic anomalies. Mineral deposits are economically profitable mineable geologic anomalies. Prospecting is the search for geologic occurrences. Exploration are all activities done to evaluate the occurrence. Exploration activities include: determination of the size, shape, grade and profit potential of the occurrence. Figure 3.1 illustrates the five stages of mine life from prospecting to reclamation. The period of a mine life is between 16 to 53 years. In Figure 3.1 the prospecting and exploration stages are highlighted. (Hartmann, 2002, p. 8)

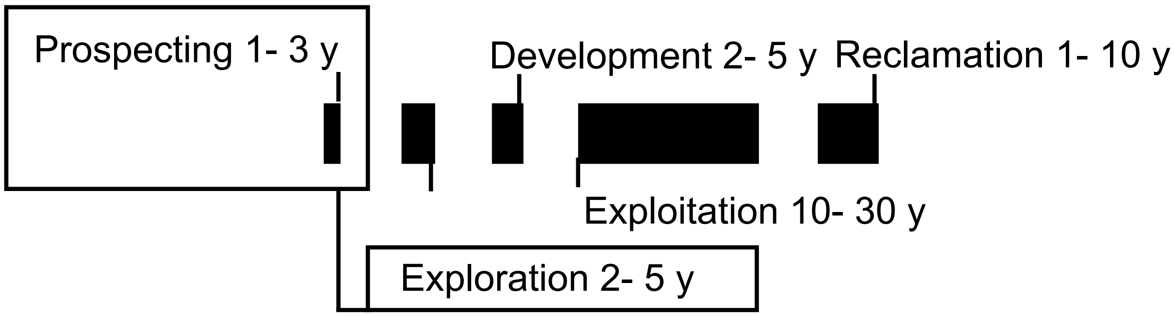


Figure 3.1 Five stages of mine life. Data from (Hartmann, 2002, p. 8)

The steps in **natural graphite exploration** are visualized in Figure 3.2. After a mineralized outcrop is discovered, field mapping and trenching is done. Outcrops of soft, weathered graphite schists are trenched. Channels are cut across un-weathered outcrops to collect rock samples. Electromagnetic surveying for the high-electric conductible natural graphite and associated metal sulphide minerals is common. The four methods aerial air borne, fixed loop, moving loop and downhole electromagnetic surveying can be conducted. To produce drilling samples two methods exist: rock chip sample drilling (inexpensive) and diamond core drilling (expensive). (Scogings a, 2015, p. 42 - 46; Scogings b, 2015, p. 79)

Part of the mineralogical and petrographic examination is the preparation and analysis of thin sections. Thin sections can be analyzed with several methods: optical or polarized-light microscopy, X-ray diffraction (XRD), scanning electron microscopy (SEM) and mineral liberation analysis (MLA). During the petrographic examination the flake size distribution is determined.* The results of the mineralogical and petrographic examination help to select methods of metallurgical testing.

During metallurgical testing mineralogical tests, crushing, assaying by size, gravity separation tests and flotation tests are performed. (Scogings a, 2015, p. 42 - 46; Scogings b, 2015, p. 79)

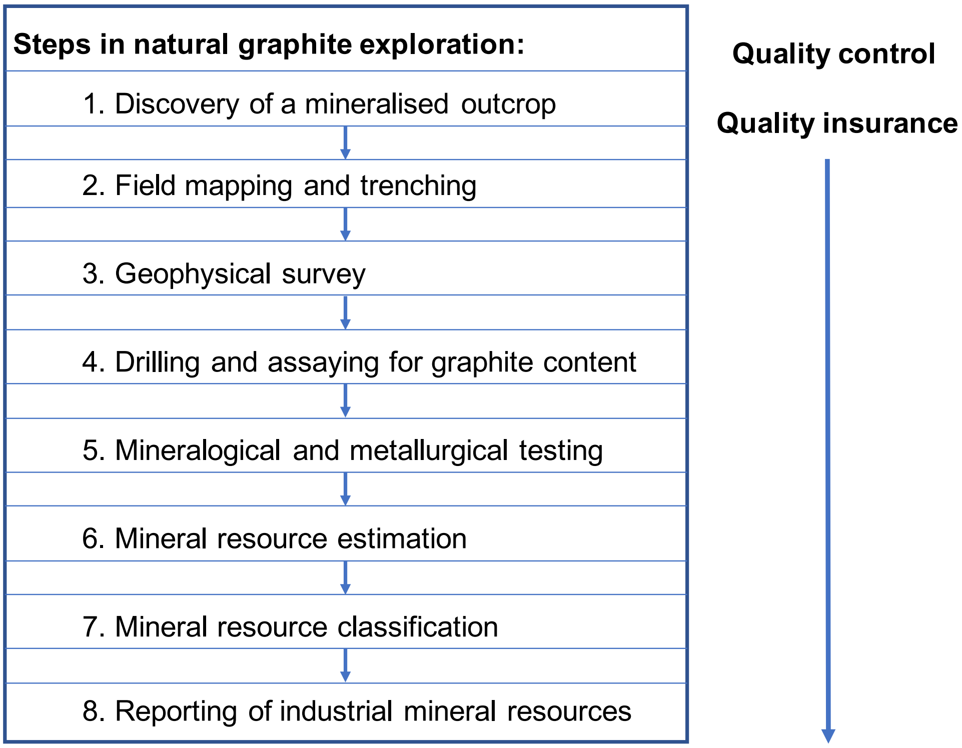


Figure 3.2 Steps: natural graphite exploration. Data from (Scogings a, 2015, p. 42- 46; Scogings b, 2015, p.79)

The reporting of industrial mineral resources is done according to international standard policies such as the Australian "JORC: Joint Ore Resource Comittee'' or the Canadian "NI 43-101'' code. The "JORC: Code'' states in Clause 49:

> For minerals that are defined by a specification, the Mineral Resource or Ore Reserve estimation must be reported in terms of the mineral or minerals on which the project is based and must include the specification of those minerals (Joint Ore Reserves Committee, 2012, p. 23).

Quality control and quality insurance must be assured during the exploration process. For example, rock chip and diamond drilling hole samples should be twinned to prevent an assay bias when soft and low density graphite from rock chip samples becomes dust. The necessity of an external checking of the samples by a third party is obvious. Figure 3.3 demonstrates examples of graphite exploration projects conducted worldwide. (Scogings a, 2015, p. 42 - 46; Scogings b, 2015, p. 79)

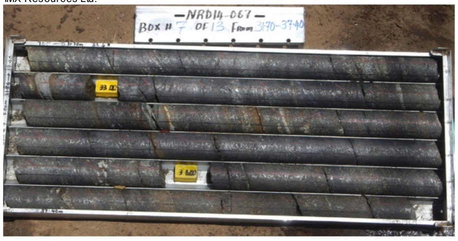
Ardiden Ltd.

Channel sampling of graphitic outcrop in Canada

Kibaran Resources Ltd.

Rock chip samples at Epanko graphite project

IMX Resources Ltd.



Diamond Drilling core samples in a core tray

Figure 3.3 Examples of graphite exploration project steps. Photographs modified from (Scogings a, 2015, p. 43)

[*The flake size distribution in the ore is an economically important factor directly related to the marketable end product price.]

3.1 Origin of graphite occurrences and deposits

Natural graphite deposits are formed by the **maturation and metamorphism*** of immature organic material and precipitation from C-O-H fluids (of metamorphic or metasomatic origin) triggered by changes in temperature and pressure, fluid buffering, or by mixing of C-O-H fluids of different compositions and probably different origins. During, the **syngenetic formation*,** carbonaceous material is dispersed in sediments (Kwiecinska, B., and Petersen, H.I, 2014, p. 104). **Precipitation*** of graphite from **C-O-H fluids** includes isobaric cooling or isothermal pressure increase. (Simandl et al., 2015, p. 168; Simandl, 1992, p. 383)

Methods to model the graphite formation process are: modeling the organic or abiotic chemical conditions (e.g. through chemical equations or diagrams), or modeling the geochemical conditions (e.g. through process schemes). A barycentric Ternary plot* is one method to model the origin of high-grade graphite deposits. Figure 3.4 is a Ternary C-O-H plot that explains the increasing size of the graphite stability field (shaded region) with decreasing temperature from 1000 °C to 400 °C. (Simandl et al., 2015, p. 168)

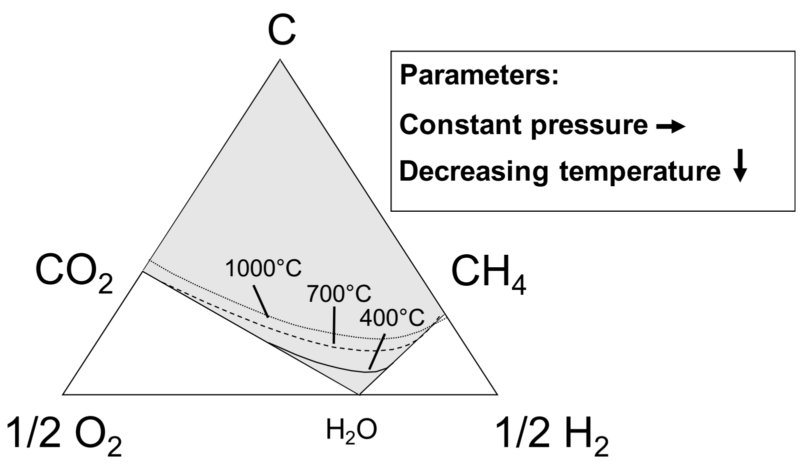


Figure 3.4 Ternary C-O-H plot of isobaric cooling. Based on data of (Simandl et al., 2015, p. 168)

Figure 3.5 is a diagram modeling the H/C to O/C atomic ratio of the carbonization* and graphitization processes. **Graphitization*** occurs at higher temperatures and higher pressures of deeper burial or metamorphism compared to the diagenesis of oil and gas. Contact or regional metamorphism may further transform organic material into an ordered crystal structure. (Simandl et al., 2015, p. 167)

[*Appendix D: Glossary]

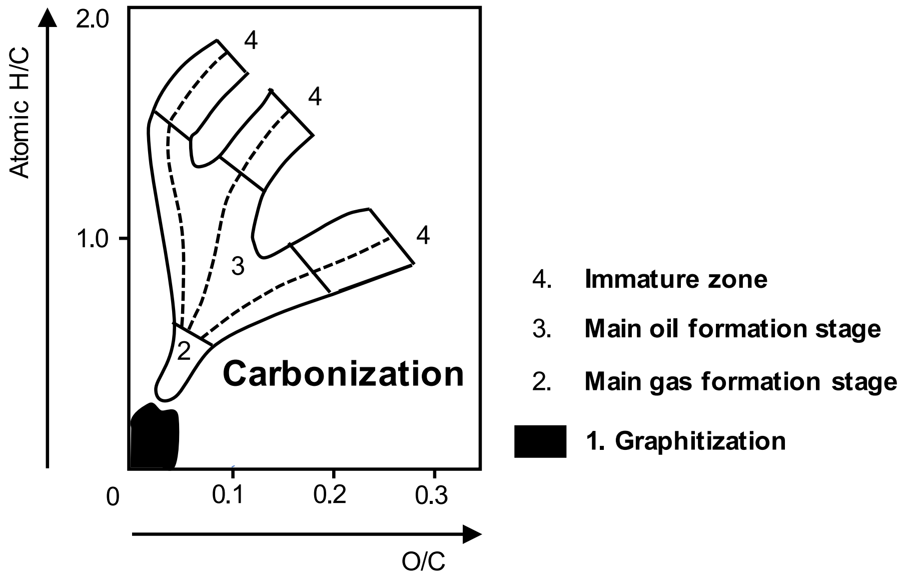


Figure 3.5 H/C and O/C atomic ratio diagram. Based on data of (Simandl et al., 2015, p. 167)

The chemical equations in Figure 3.6 demonstrate the theoretical **abiotic graphite formation process**. The chemical equations model the most probable formation process under natural conditions. The balance of CO and CO_2 plays an essential role according to this hypothesis. Figure 3.7 describes the graphite formation through **thermal dissociation of methane.**

Abiotic graphite formation

$$2\,CO \longrightarrow C + CO_2$$

$$2\,CO + 2\,H_2O \longrightarrow 2\,CO_2 + 2\,H_2$$

$$CO_2 + 2\,H_2 \longrightarrow C + 2\,H_2O$$

Figure 3.6 Chemical equations of the abiotic graphite formation. Based on data from (Kwiecinska, B., and Petersen, H.I, 2004, p. 106)

Gaseous emanations $\longrightarrow$ Methane formation: $CaCO_3 + 4H_2 \longrightarrow CH_4 + H_2O + Ca(OH)_2$

Pyrolitic dissociation of methane: $CH_4 \longrightarrow C + 2H_2$

Figure 3.7 Chemical process of graphite formation from pyrolytic methane dissociation. Based on (Kwiecinska, B., and Petersen, H.I, 2004, p. 106)

A flow scheme that explains the **geochemistry** of graphite formation from juvenile carbon is Figure 3.8. Graphite and diamond in basic and ultrabasic rocks is formed out of juvenile carbon through magmatic activity. The formation of graphite in pegmatites from juvenile carbon is not further explained. Graphite vein deposits are probably formed through metamorphism of carbonate rocks and mineralizations. Carbonate rocks and mineralizations are formed from juvenile carbon through magmatic activity of hydrothermal solutions and carbonatites. Carbonate rocks and mineralizations are also formed through the sedimentation of carbon occurring in the biosphere. Biospheric carbon is formed through the photosynthesis of atmospheric carbon dioxide. Atmospheric carbon dioxide is formed through weathering of carbonate rocks and mineralizations or through the oxidation of organic matter. Graphite in metamorphic rocks is formed through the metamorphism of bituminous matter in sediments, coal, peat or hydrocarbons. Bituminous matter is formed by the sedimentation of biospheric carbon in organic compounds and carbonates. (Krauss et al., 1989, p. 5)

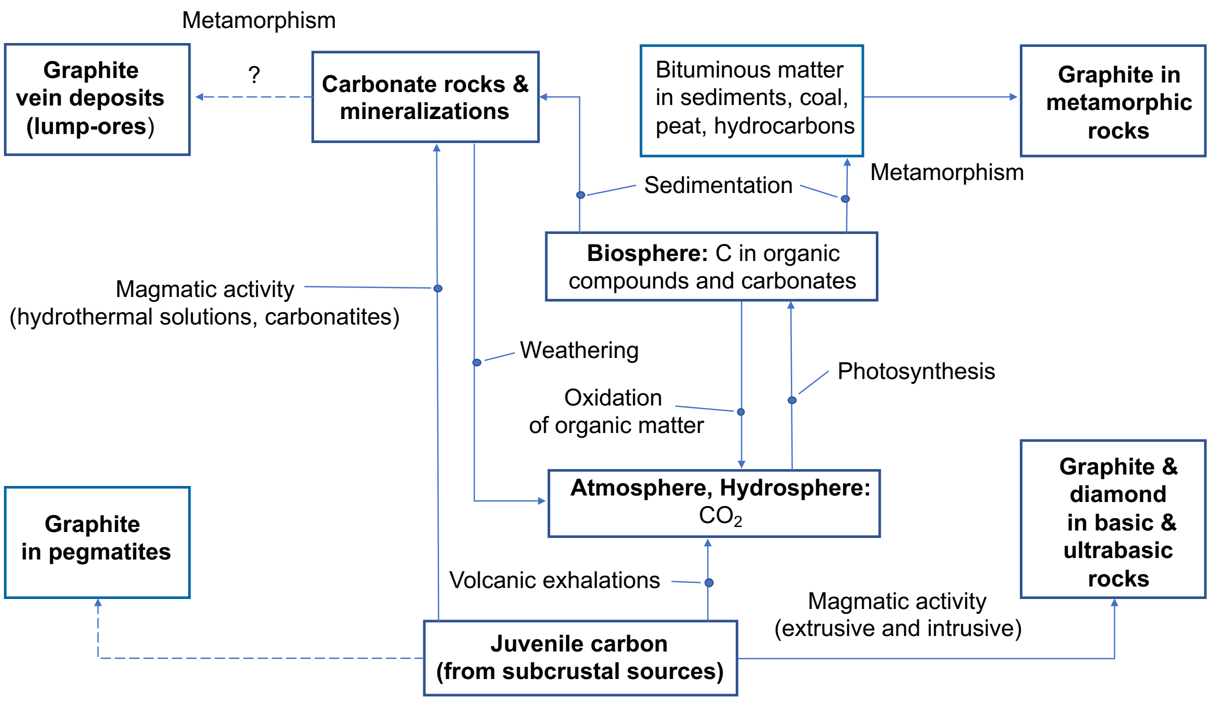


Figure 3.8 Flow scheme: Geochemistry of graphite formation from juvenile carbon. Based on data from (Krauss et al., 1989, p. 5)

The genesis of the three most significant natural graphite deposit types was reported for the first time in 1907 by Cirkel. Since 1907 numerous authors described the genesis of worldwide natural graphite deposits. Natural graphite particles in occurrences and deposits are distributed throughout a **host rock**. A brief overview of the genesis and characteristics of vein-type graphite deposits, amorphous/ microcrystalline graphite deposits and crystalline flake type graphite deposits:

- **Genesis and characteristics** of **vein-type** graphite **deposits:**

Vein-type natural graphite deposits are found in metasedimentary belts metamorphosed to amphibolite and granulite facies. In vein-type natural graphite deposits chips of size greater than 300 µm occur. Vein-type natural graphite is formed in veins or accumulations along intrusive pegmatites with limestones. Natural graphite metamorphosed in spaces and facies is currently inter alia mined in Sri Lanka and Mexico. (Taylor, 2006, p. 507)

Simandl et al. explain details of the origin of vein-type graphite deposits:

> The source of carbon for vein - type deposits varies (Simandl et al., 1997, p. 2). In [metasedimentary] belts vein graphite deposits are found in skarn-type assemblages adjacent to igneous intrusions, in igneous intrusions, and in zones with a retrograde overprint […] (Simandl, 1992, p. 383). Other vein type graphite deposits are formed in pods and lenses, irregular bodies, stockworks, and saddle reefs (Simandl,1992, p. 383; Simandl and Keenan, 1998, p. 1). Rosettes, coarse flakes, fibers or needles oblique or perpendicular to wall rock and, in some cases, schistosity subparallel to the vein walls are characteristic textures. Outside of upper amphibolite to granulite facies metamorphic terrains and related intrusives (e.g., Cirkel, 1907), graphite veins, breccias, and stockworks also cut a variety of mafic and ultramafic rocks (e.g., Stress, 1965; Barrenechea et al., 1997; Crespo et al., 2006). (Simandl et al., p. 165)

- **Genesis and characteristics** of **amorphous/microcrystalline** graphite **deposits:**

Amorphous graphite/microcrystalline graphite particles are distributed in weakly metamorphosed rocks. Microcrystalline graphite particles range in size from 75 - 110 µm. The graphite content in amorphous deposits is dependent on the carbon amount present in the original sediments. (Taylor, p. 507)

Taylor explains the origin and consistency of amorphous deposits:

> Most microcrystalline amorphous graphite deposits are formed by subgreenschist to greenschist contact metamorphism or regional metamorphism of coal seams (Taylor, 2006). Microcrystalline deposits consist mainly of small graphite particles inter grown with impurities. Typical deposits are stratiform or lens-shaped; beds may be deformed and/or represented by folding and faulting. Pinching and swelling of beds is common. Deposits [may] consist of serval beds, each up to a few meters thick. They [may] be exposed for hundreds of meters along strike. The ore contains from 30 to 95 % graphite and, in many cases, more than 80 % (Taylor, 2006). (Taylor, 2006, p. 507 - 509)

- **Genesis and characteristics** of **crystalline flake type** graphite **deposits**:

The most common host rocks for crystalline flake deposits are paragneiss and marble that have been metamorphosed to upper amphibolite and granulite facies. Crystalline graphite particles range in size from 110 - 300 µm.

Simandl et al. explain the consistency of crystalline flake deposits:

> Disseminated graphite flakes are in a variety of rocks including marble, paragneiss, iron formation, quartzite, pegmatite, syenite (Simandl, 1992; Simandl et al., 1995) and, in extremely rare cases, serpentinized ultramafic rocks (e.g., Crespo et al., 2006). [..] Graphite deposits consisting of thick sequences of paragneiss are evenly mineralized and generally grade 2-3 % graphite or less. [...] The highest graphite grades in paragneiss-hosted deposits are along or near paragneiss-marble contacts [...] marble is separated from biotite gneiss by calcsilicate rocks (clinopyroxenites) and graphite- bearing scapolite paragneiss [...] The contact between this graphite-rich unit and the biotite-gneiss is gradational and graphite content decreases with increasing distance from the calcsilicate rocks. [...] For some deposits [...] the highest grade graphite is encountered in the crests of folds and is accompanied by retrograde minerals such as epidote and chlorite (Marchildon et al., 1993). Marbles in terrains metamorphosed to granulite facies display a granoblastic texture and generally contain less 0.5 % crystalline flake graphite, although concentrations from 1 to 3 % crystalline graphite are common. Graphite is regularly distributed throughout the host rock and the size of graphite flakes and calcite or dolomite crystals is directly correlated. Microscopic signs of corrosions or overgrowth on the graphite flakes that would indicate disequilibrium are lacking. Minor constituents such as diopside, magnesite, quartz, tremolite, forsterite, humite group minerals, garnets, scapolite, wollastonite, feldspar, phlogopite, muscovite, and serpentine account for less than 5 % per volume of the rock. Marbles with porphyroblastic texture are unusual. They contain from trace to 25 % crystalline flake graphite. (Simandl et al., 2015, p. 165)

The kinetics of crystalline flake graphite formation is influenced by the nature of hydrocarbon precursors, the partial pressure of CO_2, CO, CH_4, H_2O and H_2 and the regional pressure temperature conditions. Flake graphite is formed by the graphitization of existing aromatic C-H rings. Aliphatic C-H strings are not easily graphitized. Figure 3.9 depicts graphitized, aromatic C-H rings and non graphitized, amorphous C-H strings. (Gautneb et al., 2000, p. 73)

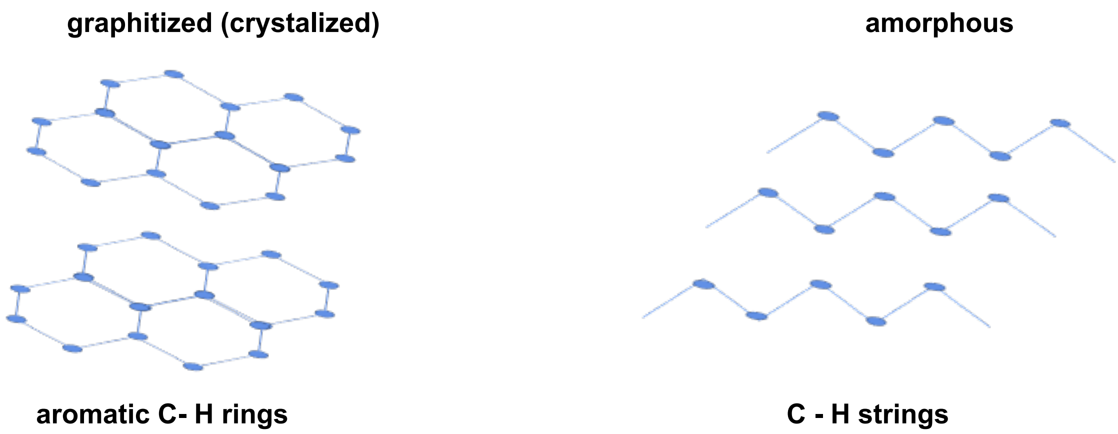


Figure 3.9 Schematic model of flake graphite formation

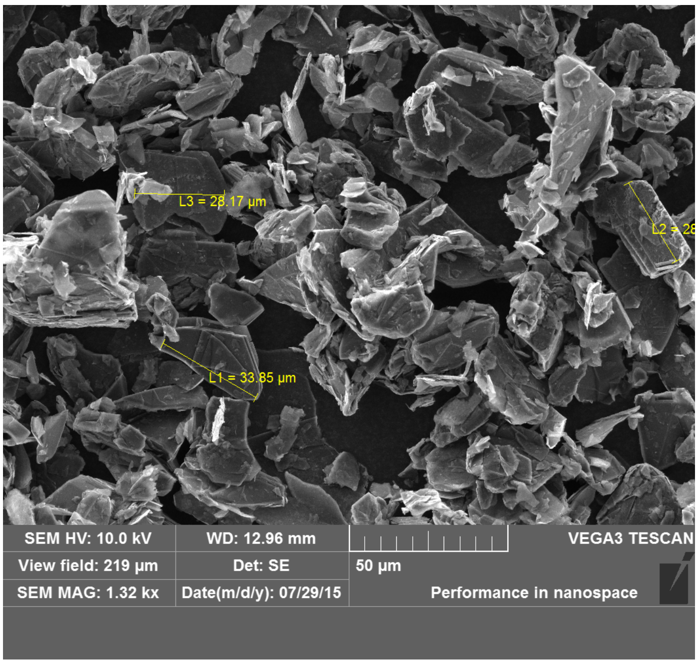


Figure 3.10 Scanning electron microscopy (SEM) of flake graphite (Great Lakes Graphite Corp., 2017)

Figure 3.10 is a scanning electron microscopy of crystalline graphite flakes sized between 35 µm to 25 µm taken in July 2015 at the laboratories of the Great Lakes Graphite Corporation. The flakes have basal cleavage and are opaque. (Great Lakes Graphite Corp., 2017; Taylor, 2006, p. 508)

3.2 Occurrences and deposits in the European Union

Figure 3.11 illustrates the location of the European natural graphite deposits most mentioned: Kropfmühl (Passau, Germany), Kaiserberg (Leoben, Austria), Woxna (Edsbyn, Sweden), Nunasvaara (Kiruna, Sweden), Skaland (Lofoten - Norway), Zavalyevsky (Odessa, Ukraine). (Industrial Minerals a, 2017, p. 1; Krüger et al., 2017, p. 125)

Figure 3.11 Map main natural graphite deposits in Europe. Based on data from: (Industrial Minerals a, 2017, p. 1; Krüger et al., 2017, p. 125)

Europe’s most important natural graphite occurrences and deposits are found in the northern countries. Gautneb states: „The Fennoscandian graphite and Russian shungite occurrences are all of paleo- or Mesoproterozoic age [*].“[Fahl] (Gautneb, 2016, p. 3)

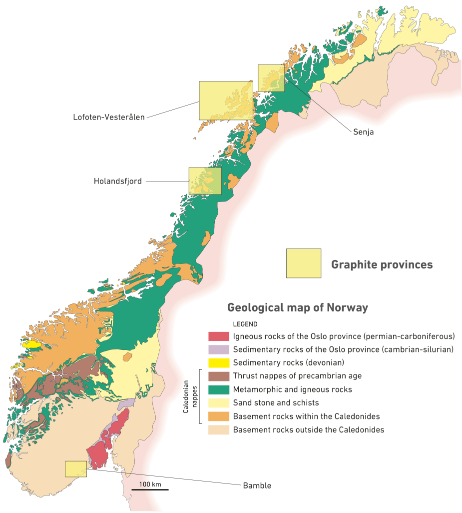


Figure 3.12 Geological map of Norway (Gautneb, 2015, p. 1)

[*Appendix B: illustrates the geotimes]

In **Norway** there are four graphite provinces: the island of Senja, Lofoten-Vesteralen, Holandsfjord and Bamble (Figure 3.12). (Gautneb, 2015, p. 1) Havard Gautneb describes the Norwegian graphite occurrences:

> All the Norwegian graphite occurrences are of flake graphite type and occur in Proterozoic supracrustal rocks of high amfibolite or granulite facies metamorphic conditions. (Gautneb, 2015, p. 1)

The **Skaland** graphite **mine** is located adjacent to the Ånderdalen Nationalpark on Senja- Island in northern Norway. The deposit consists of lenses of graphitic rock enclosed in mica schists with an ore content of around 25 % - 30 % graphitic carbon. (Gautneb et al., 2016, p. 13; Taylor, 2006, p. 511)

The **Nunasvaara** metamorphic graphite occurrence in northern Sweden consists of disseminated to massive graphite flakes. Lynch characterizes the occurrence mineralization:

> [The flakes are within] a schist horizon that forms part of a relatively conformable, polydeformed Paleoproterozoic greenstone succession (basalts, tuffs, doleritic sills, intercalated sedimentary rocks). [Fahl] (Lynch et al., 2016)

The **Kropfmühl mine** is located in South Germany, Bavaria, Passau. The graphite bearing host rock is gneissic and schistose rock of the Bohemian massif. The graphite layer mined is 1.5 m thick. The ore content is according to the company: Graphitwerk Kropfmühl AG. between 20 % - 25 % graphitic carbon. (Taylor, 2006, p. 513)

The underground mine **Kaiserberg** is located in Austria, Styria, Leoben. The metamorphic deposit was formed during Paleozoic. The graphitic beds are enclosed in graphitic schists and quartzites. The ore content is between 30 % - 80 % graphitic carbon. (USGS b, 2017, p. 1; Taylor, 2006, p. 511) Taylor defines the deposit structure:

> […] these Alpine deposits are embedded in a zone of fine - grained, scaly schists in lenses and individual beds as much as 10 m thick. The ore is black, soft, fine - grained, and dense, with a carbon content of 40 % - 88 %. It is low in sulfur (0.2 % - 0.3 %) and iron and practically free of carbonate and phosphate. (Taylor, 2006, p. 512)

The **Zavalyevsky complex,** Ukraine, Odessa contains graphite, quartzite, calciphyre, gneiss and calc-silicate hornfels rocks. Other deposits consisting of flake graphite disseminated in mica schist or mica gneiss are found in the **Czech Republic.** (Buzilo et al., 2011, p. 19; Taylor, 2006, p. 511)

3.3 Example: Woxna deposit complex in Sweden

The Woxna deposit complex is an interesting example regarding a future independent European natural graphite supply. The Woxna deposit complex is located near the village of Edsbyn in central Sweden. The complex consists of the four deposits Kringelgruvan, Gropabo, Mattsmyra and Månsberg. A voluminous exploration documentation over the Woxna deposit complex exists (see Appendix C).

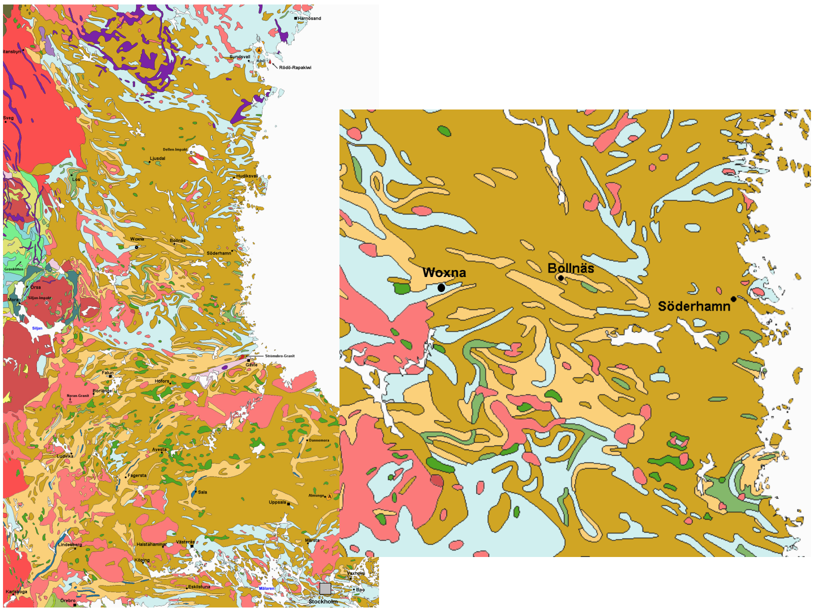


Figure 3.13 Section: Map bedrock of Sweden, Scale 1 : 1 250 000 (Geological Survey of Sweden (SGU), 2007)

Figure 3.13 is a geological map of the bedrock of Sweden in the scale 1 : 1 250 000. The bedrock is a part of the “Fennoscandian Shield'', a stable rock area consisting of Precambrian rocks. The Precambrian period lasted from 4,600 million years ago until the Cambrian period 545 million years ago. Sedimentary rocks rest upon the Precambrian shield area. The Woxna deposit is part of the trans-scandinavian magnetite belt**.** According to the map “bedrock of Sweden'' the region around Woxna consists of sandstone and conglomerates. (Geological Survey of Sweden (SGU), 2007)

Reed summarizes the **local geological setting** and **mineralization** of the four deposits:

> The Kringelgruvan claim shows development of trace to massive graphite in meta sedimentary and metavolcanic host rocks which have been metamorphosed to sillimanite grade and intruded by felsic units ranging from alkali pegmatite to granite.
>
> At Kringelgruvan, the geology is dominated by steeply-dipping, calcareous quartz- rich meta-tuff, with interbedded metasedimentary units and cross-cutting pegmatite.
>
> The mineralization is tabular in shape, and late in the structural history, postdating and cross-cutting any remnant tectonised and metamorphosed lithologies.
>
> At Mattsmyra and Gropabo, the local geology is dominated by steeply to moderately dipping porphyroblastic metavolcanic and meta-argilic rocks with common intrusive alkali pegmatites.
>
> Graphite mineralization occurs in prehnite-bearing meta-tuffs, garnetiferous meta-argillites and pegmatitic gneiss in at least three discontinuous, stratiform graphite-pyrrhotite horizons. (Reed, 2015, p. 7)

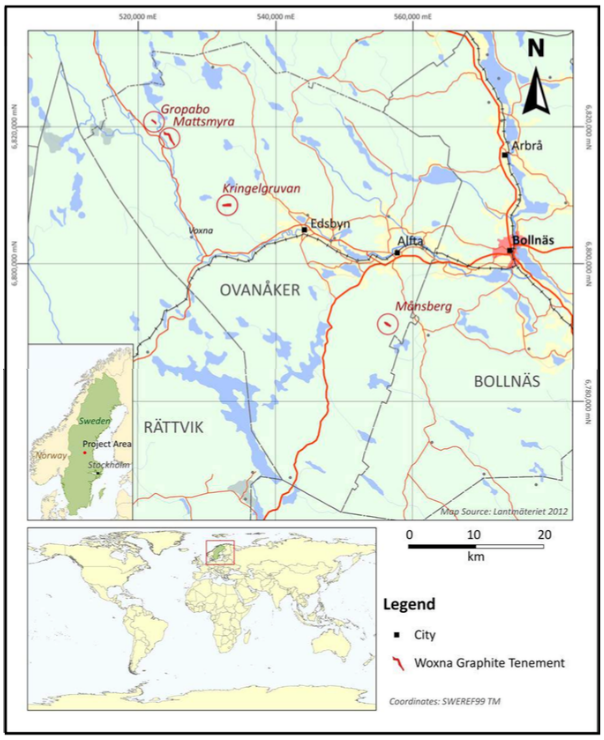


Figure 3.14 Map: "Woxna Graphite AB" graphite properties (Reed, 2015, p. 16)

Figure 3.14 illustrates the location of the four graphite deposits: Kringelgruvan, Mattsmyra, Gropabo and Månsberg. Table 3.1 lists the chemical composition of the Woxna graphite ore analyzed during an experimental study of Woxna fine concentrate. The mineral composition is: graphite 87.5 %, clays 5.5 %, mica 4.0 %, feldspar and quartz 1.5 %, pyrrhotite 1.5 %. (Lu et al., 2002, p. 756)

Table 3.1 Chemical properties: Woxna fine graphite concentrate. Data from (Lu et al., 2002, p. 756)

Mineralogical properties		**Woxna deposit**	
Compound	Amount	Compound	Amount
C	87.7 %	MnO	0.02 %
S	0.6 %	K_2O	0.65 %
SiO_2	5.25 %	Na_2O	0.06 %
Al_2O_3	2.93 %	P_2O_5	0.01 %
Fe_2O_3	1.65 %	TiO_2	0.13 %
CaO	0.35 %	Trace elements	0.02 %
MgO	0.55 %		

Appendix C gives an overview of the prospection and exploration reports conducted over the Woxna deposit. The deposit was first prospected in 1983. Five drilling and exploration reports, three technical beneficiation (leaching) studies and six technical reports were conducted until 2015 (Figure 3.15). * (Reed, 2015, p. 117 - 118)

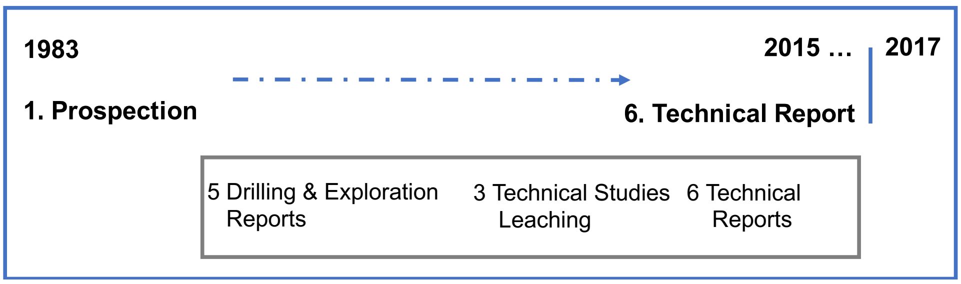


Figure 3.15 Timeline 1. Prospection to 6. Technical Report. Data from (Reed, 2015, p. 117 - 118)

* [Personal comment: The future development of the "Woxna project'' is questionable even after 32 years of geological surveying. Two years after publishing of the last technical report, still no mine construction activities are started. [Fahl]]

4 Technical Process: Extraction

Mineral resources are usually extracted by underground or surface mining. The mining method depends on the locational, natural and geologic factors. Locational factors include labor availability, support service, operational impacts of climate and weather. The geologic factors include criteria such as topography, spatial relations of the ore body, mineralogy, petrography and rock mechanics properties. The Figure 4.1 illustrates the five stages of mine life from prospecting to reclamation. The period of a mine life is between 16 to 53 years. The development and exploitation stage is highlighted. (Hartmann, 2002, p. 8, p. 98 - 100)

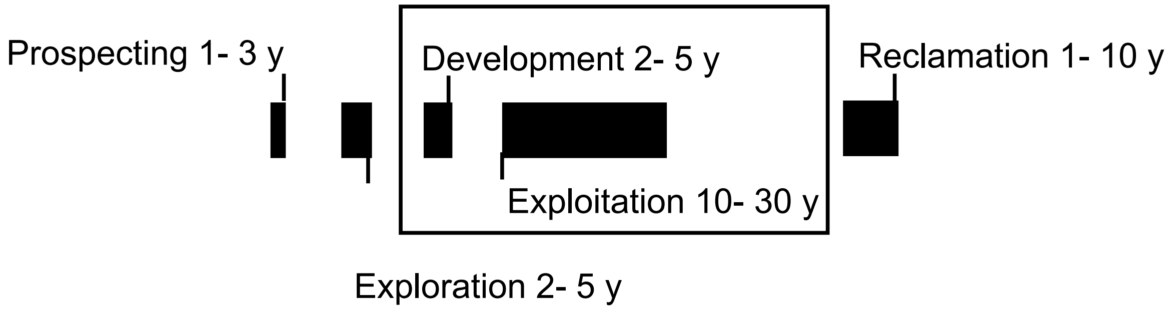


Figure 4.1 Five stages of mine life. Data from (Hartmann, 2002, p. 8)

Natural graphite is mainly mined in five countries: China, India, Brazil, Canada and North Korea. 69 percent of the worldwide natural graphite production is extracted in China. Figure 4.2 demonstrates the relation of the worldwide graphite production by country. The major graphite production regions are depicted.

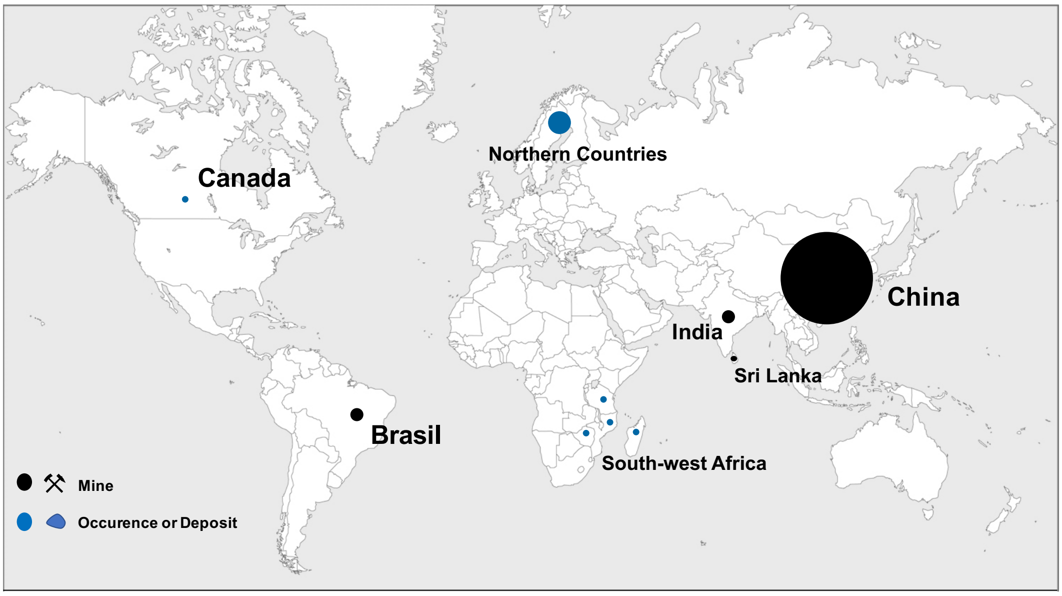


Figure 4.2 Map: worldwide graphite regions. Data from (Industrial Minerals a, 2017; USGS a, 2017, p. 75)

4.1 Process scheme: Extraction

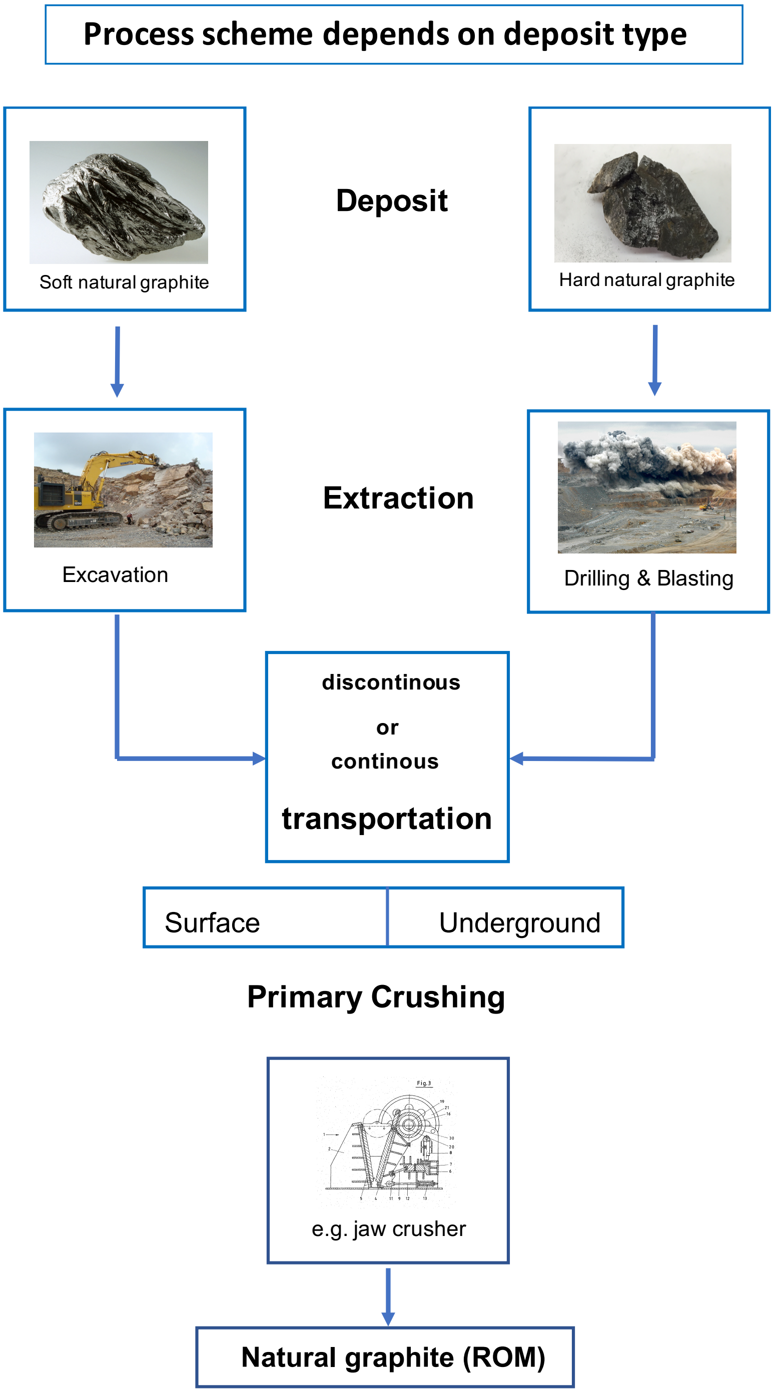


Figure 4.3 General process scheme natural graphite extraction (Fahl, 2017)

Figure 4.3 is a general schematic description of the graphite mining process. According to the scheme: the process depends mostly on the deposit type and hardness of the natural graphite ore. The main three process steps are extraction, transportation and primary crushing.

4.2 General process description

The stages of underground mine planning and mine development are depicted in Figure 4.4. Primary development includes the construction of the main mine openings: e.g. construction of slopes, ramps or shaft sinking. Aim of the primary development is to create a primary access to the deposit. Secondary development includes all activities that prepare the deposit for extraction: e.g roadheading, installation of appropriate mine ventilation, installation of energy and water supply. The term tertiary development describes all operation measures done during ore excavation: e.g. drilling and blasting, waste air management, loading and hauling, rock support and surveying. Long-term mine planning includes scheduling of operations to do the primary and secondary development. Short-term operational planning schedules weekly or shift-based activities such as drilling & blasting patterns. Modern software is used to support the scheduling. (Hartmann, 2002, p. 268)

Stages of underground mine planning and development

Operation & drivage			Mine planning & design
Primary development	Construction primary infrastructure Aim: primary **access to deposit**	e.g. shaft sinking	**Long-term** planning
Secondary development	Construction secondary infrastructure Aim: **preparation** of deposit **for mining**	e.g. roadheading	
Tertiary development	**Direct excavation method**	e.g. stoping	**Short-term** operational planning

Excavation method generally depends on deposit type !

Figure 4.4 Stages of underground mine planning and development

The stages of surface mine planning and design are depicted in Figure 4.5. During pit design and operation of the mine the aim is to solve the trade-off between economic parameters, technical limits and health & safety and law regulations. Economic parameters are for example: the defined cut-off grade, the mineral price and the aspired production rate. Technical limits are for example: the equipment supply and the maintenance rate and quality. Safety limits are for example: rock mechanics, water management and slope design. (Hartmann, 2002, p. 153 - 176)

Stages of surface mine planning and development

Development & construction

Pit Layout Planning
Construction surface facilities
Pre-production stripping
Advanced stripping
Extraction according to mining method and equipment selection
Reclamation

Equipment selection generally depends on ore & deposit type !

Figure 4.5 Stages of surface mine planning and development

Figure 4.6 gives a synopsis over the mining methods commonly applied on the natural graphite types: vein, crystalline and amorphous. Figure 4.6 also outlines the geological and mineralogical deposit features. Compared to other critical raw materials (e.g Rare Earth Elements), natural graphite is not mined as a by-product.

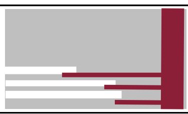
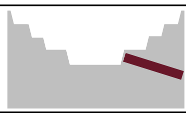
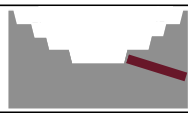
Natural graphite type	Vein-type	Crystalline	amorphous
Mining method	underground	surface	surface
			
Main Geological features	Igneous intrusions Irregular bodies	Disseminated flakes in a host rock (marble or paragneis)	Stratiform or lens shaped Folded beds
Deposit geometry	Veins and other graphite filled open spaces form pods and lenses, irregular stockworks, bodies, saddle reefs and breccias	Thick sequences of paragneisses and marbles. Highest grade near paragneiss-marble contacts and in folds.	Stratiform or lens shaped, folded and faulted, pinching and swelling of beds is also common
Main host rocks	Marble, paragneiss, iron formation, quartzite, pegmatite, syenite, and in rare cases mafic	Marble, paragneiss, iron formation, quartzite, pegmatite, syenite and extremely rare cases ultramafic rocks	Chlorite and muscovite schists, phyllites, quartzites, metagraywakes, limestones, sandstones and conglomerates
Metamorphic grade	Upper amphibolite to granulite facies, in skarn-type assemblages adjacent to igenous intrusions, in igenous intrusions, and in zones with a retrograde overprint	Upper amphibolite to granulite facies	Sub-greenschist to greenschist contact, or regional
Ore characteristics	90 % graphitic-carbon. Rossettes, coarse flakes, fibers or needles oblique or perpendicular to wall rock. Sometimes, schistosity subparallel to the vein walls	Grade from < 2- 15 % graphite; Paragneisses evenly mineralized grade < 3 % graphitic carbon Marble (granoblastic) contain < 0.5 – 3 % graphite	30 - 90 % graphite, in many cases > 80 %, mainly consits of small graphite Particles intergrown with impurities

Figure 4.6 Synopsis mining methods for natural graphite types and geological features. Based on data from (Simandl et al., 2016, p. 1)

Vein-type natural graphite is mined underground in Sri Lanka, Mexico, the United States and India. Production from vein-type natural graphite mines accumulated to 1 % of the worldwide natural graphite production in 2012. Due to the high carbon content of 90 - 95 % underground mining is economically feasible. The vein-type graphite deposits in Sri Lanka are located in Southern, Western and Sabaragamuwa provinces. The thickness of the veins commonly range from a few millimeters to almost a meter. Table 4.1 lists the associated minerals reported in Sri Lankan deposits. (Taylor, 2006, p. 512; Industrial Minerals b, 2017, p. 1)

Table 4.1 Associated minerals in Sri Lankan deposits with data from (Taylor, 2006, p. 512)

Associated minerals in Sri Lankan deposits		
quartz	apatite	allanite
pyrite	pyroxene	magnetite
calcite	rutile	lime-magnesia silicates

Two graphite mines operate in Sri Lanka. According to Kahatagaha Graphite Lanka Ltd., owned by the Gouvernement of Sri Lanka: 80 tonnes natural graphite with a minimum purity of 90 % carbon content were mined in 2013. The production level is operated in a depth of 350 m. Bogala Graphite Ltd., produces 250 tonnes monthly at an underground level of 700 m depth from one graphite vein. (Plumbago Refining Corp., 2013, p. 1; Taylor, 2006, p. 512)

Crystalline flake natural graphite is mined in surface mines. Crystalline flake is the economically most important natural graphite type. Production from crystalline flake natural graphite mines accumulated to 55 % of the worldwide natural graphite production in 2012**.** The current common cut-off grade defined for crystalline flake mines is 3 - 5 % ore content. The common ore content in crystalline flake mines varies between 3 - 20 %. Figure 4.7 shows the percentages of crystalline flake graphite produced worldwide by country. China produced in 2012 the main share of 66 % of the crystalline flake graphite production. Advanced exploration projects exist in Mozambique, Madagascar and Canada. (Desautels, 2014, p. 3, p. 6; Scogings b, 2015, p. 78; Industrial Minerals b, 2017, p. 1)

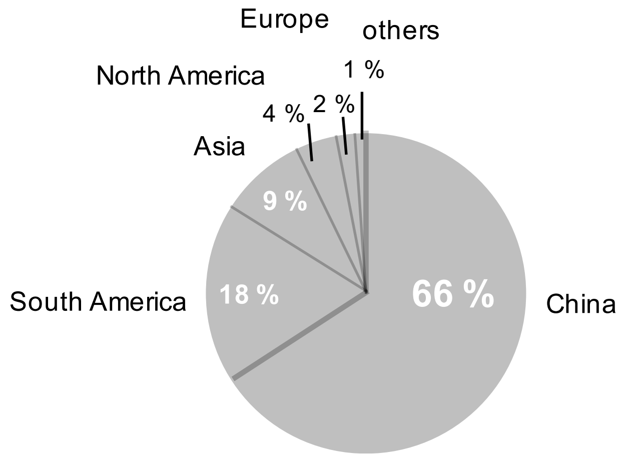


Figure 4.7 Worldwide flake graphite production in 2012. Data from (Industrial Minerals b, 2012, p. 1)

Amorphous natural graphite is mostly mined in surface mines. Production from amorphous natural graphite mines accumulated to 44 % of the worldwide natural graphite production in 2012. Figure 4.8 shows the percentages of amorphous natural graphite produced worldwide by country. China produced in 2012 the main share of 89 % of the amorphous natural graphite production. Amorphous natural graphite dispersed in chlorite-sericite schist ore is mined underground at Kaiserberg mine Austria, Styria, Leoben. Amorphous natural graphite can only be used in low-value applications. (Taylor, 2006, p. 507; Simandl et al., 2016, p. 1; Industrial Minerals b, 2017, p.1)

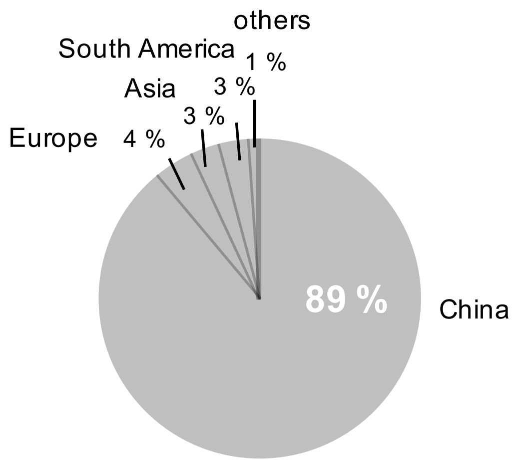


Figure 4.8 Worldwide amorphous graphite production in 2012. Data from (Industrial Minerals b, 2012, p. 1)

Table 4.2 Overview: Important natural graphite mines. Data from (Industrial Minerals a, 2017, p. 1)

Country	Mine(s)	Mining method	Status	Graphite types
Austria	Kaiserberg	underground	inactive	amorphous
Brazil	Nacional de grafite	underground	active	vein-type
Canada	several different	surface	active	crystalline flake
China	several different	artisanal mining, surface & underground	active	crystalline flake
Germany	Kropfmühle	underground	inactive	vein-type
India	several different	surface & underground	active	mixed
Madagascar	Loharano	underground	development	crystalline flake
Zimbabwe	Lynx	underground	development	crystalline flake

Table 4.2 lists important natural graphite mines worldwide. It evaluates the current production status, the location and geologic deposit type of the mines. Natural graphite mines are small-scale mining projects concerning operating and investment costs. Most of the active, producing natural graphite mines operate in China. The annual Chinese graphite production accumulates to 780,000 tonnes per year. Approximately 600,000 tonnes per year are produced in artisanal mining operations. In Brazil two underground vein-type mines operate and produce 80,000 tonnes of natural graphite per year. In India 10 mining districts exist and produce an accumulated production of 170,000 tonnes per year. The most important district is the Odisha region. The typical ore content of producing Indian open-pit mines is between 5 - 20 % graphitic carbon. Indian open-pit mines operate close to surface (maximal depth 45 m). Underground mines existing in India operate at greater depths. An example is the Sargipalli underground mine with an ore content of up to 40 % graphitic carbon. The major technical problem of the Indian Mining Industry is dewatering. Canada is a leading country for the development of new natural graphite mines. Eight companies have acquired exploration licenses for different properties. (Industrial Minerals a, 2017, p.1; Indian Bureau of Mines, 2016, p. 5; USGS, 2017 a, p. 75)

4.3 Case studies: Lac Knife Mine & Jongchon Mine

Table 4.3 Data sheet "Jongchon Mine". Based on data from (Olson 2016, p. 74; Choi 2010, p. 28)

Mine features	
Jongchon **Geographical location:** (정촌광산)	37°55'7.23″N,126°6'49.34″E Jongchon-ri, Yonan County, South Hwanghae, North Korea
Figure 4.9 Satellite picture "Jongchon Mine" (Google a, 2017)	
Figure 4.10 Map geographical location: "Jongchon Mine" (Google b, 2017)	
Yearly production capacity [t ore]:	75,000
Yearly production (2009) [t ore]:	30,000 - 40,000
Equipment	
Excavation - Hydraulic Excavator, Shovel Volume 3 m^3	Number: 1
Loading - Hydraulic Excavator, Shovel Volume 3 m^3	Number: 1
Transport - Truck, payload 20 t	Number: 1

Beneficiation technique: Flotation
Marketable product: planned production (2009) 3,000 t/a graphite concentrate 98 % C, realistic production 1,500 t/a graphite concentrate 90 % C
Special features of the mine: Cooperation project between North and South Korea Insufficient production reason → insufficient energy supply

Table 4.3 is a data sheet for the "Jongchon Mine". Figure 4.9 is satellite picture of the mine. Figure 4.10 shows the location of the mine on a North Korean map.

The "Jongchon Mine" is a surface mine located Yon-an area of South Hwanghae Province in south-west North Korea (Figure 4.10). The mine is supplied with energy by Pyeongchang coal plant. The planned production capacity of 3,000 t/a marketable graphite concentrate with 98 % carbon content was not reached due to insufficient energy supply. A realistic estimation for the current production rate is 1,500 t/a marketable graphite concentrate with 90 % carbon content. The average ore grade is 5.53 % graphic carbon. The "Jongchon Mine" is an inter-korean development project. The mining operation started in April 2006. The development costs of US$ 10.2 million were equally shared by the cooperation partners Myongji Corp. (North Korea) and Korea Resources Corp. (Republic of North Korea). In 2006 during the test run the mine was temporarily shut down for the first time due to insufficient water and energy supply. In 2008 the mining operation was reopened. (Wu, 2007, p. 3; Olson 2016, p. ; Choi 2010, p. 28)

Concerning the equipment and operations costs the "Jongchon Mine" is a small scale mining project. The equipment fleet consists of one hydraulic backhoe excavator with 3 m^3 shovel volume and of one truck with 20 t payload. The hydraulic backhoe excavator is used for ore extraction and self loading of the truck. (Olson 2016, p. 74; Choi 2010, p. 28)

Table 4.4 Data sheet “Lac Knife” exploration project. Data from (Desautels et al., 2014, p. 155 - 164)

Exploration project

Lac Knife	**Geographical location:**	52°33’N, 67°11’W Fermont, Quebec, Canada

Figure 4.11 Aerial picture “Lac Knife area” (Focus Graphite Inc., 2015)

Figure 4.12 Map geographical location “Lac Knife area” (Google c, 2017)

Planned yearly production capacity [t ROM ore]:	313,470
Yearly production [t ore]:	None
Total marketable graphite production [dry t per year]:	44,300

Equipment

Excavation - production drill rigg (e.g. MD5125), borehole diameter: 114 m	Number: 1
Loading - Hydraulic Excavator (e.g. 390D), Shovel Volume 4 m^3	Number: 1
Transport - Trucks (e.g 770G), payload 36.5 t	Number: 4
Other - Wheel loader, Shovel Volume 6 m^3	Number: 1

Beneficiation technique: Flotation
Purification technique: thermal halogenation, chlorination
Marketable product: planned production: 44,300 t/a graphite concentrate 98 % C, planned production: spherical graphite concentrate 99.98 % C (no mass data)
Special features of the mine: Mine is operated by a contract company, seasonal production in summer, production from ore stockpile in winter
Estimated production costs: US$126.95 per tonne concentrate

Table 4.4 is a data sheet about the "Lac Knife" exploration project. The "Lac Knife" exploration project is a Greenfield exploration project located at Lac-des-Iles in the Côte Nord region of Québec, Canada. Figure 4.11 is an aerial picture of the property. On the aerial picture the beginning of the stripping operation is visible. Figure 4.12 shows the location of the "Lac Knife" exploration project on a Canadian map section.

Taylor summarizes the deposit geology:

> The host rock is a jumble of Grenville biotite-garnet gneiss, quartzite, and graphite-containing marble. Although most of the graphite is found in the higher-grade marble core of a gneissic body, some larger-flake graphite is found in the quartzite ring surrounding the core. The entire sequence is cut by gabbros-diorite dikes and pegmatite bodies and is extensively folded and faulted.
>
> (Taylor, 2006, p. 511)

According to the "NI 43-101 Technical report" prepared on the feasibility study: a conventional open pit, truck and shovel, drilling and blasting, mining operation is planned. The estimated end depth is 100 m. In the feasibility study a starter pit is designed to optimize the stripping ratio in the first five years. The "NI 43-101 Technical Report" prepared on the feasibility study describes the following scenario: The mine operation is run by a contract company to save operating costs. The ore is extracted by drilling and blasting. A hydraulic backhoe excavator with 4 m³ shovel volume is used for ore loading. The ore is transported with four trucks with a payload of 36.5 t. The mining operation is run during six summer months and the ore is stockpiled. The overburden removal is done in winter to take advantage of the frozen ground conditions. In winter the beneficiation plant is feed from this stockpile by a wheel loader with 6 m³ shovel volume. The mining operation is supplied with energy from the Hydro-Québec power line passing five kilometers east of the project. (Desautels et al., 2014, p. 1, p. 155 - 168)

The estimated mining costs stated in the feasibility study are US$126.95 per tonne concentrate. The estimated life of mine operating costs stated in the feasibility study are US$444.1 per tonne concentrate. The average ore grade stated in the feasibility study is between 14 - 17 % carbon content. The Focus Graphite Incorporation plans the production of marketable spherical graphite concentrate with 99,98 % carbon content. The planned purification process is thermal halogenation and chlorination. The planned annually production capacity is 44,300 t/a graphite concentrate with 98 % carbon content. Figure 4.13 is a draft of the "Lac Knife'' mine plan. The draft shows the geological features and the location of the planned mine facilities and the tailings pond. (Desautels et al., 2014, p.7, p. 13 - 14)

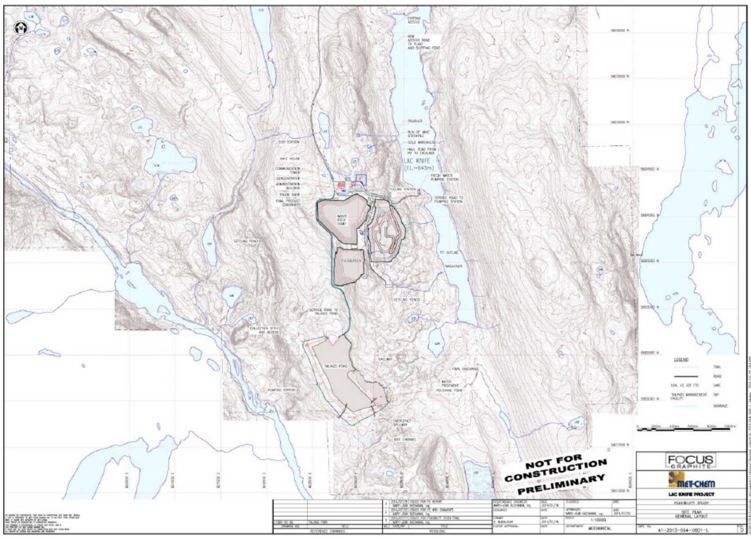


Figure 4.13 Draft of a mine plan from the "Lac Knife'' exploration project designed by Met - Chem for Focus Graphite Inc. (Focus Graphite Inc., 2015)

5 Technical Process: Beneficiation

Mineral resources are mechanically treated without changing their physical properties during beneficiation. Beneficiation is economically feasible if the price increase of the product is higher than the beneficiation costs. The quality of the beneficiation process is essential for the rentability of the company. Compared to mining, beneficiation has higher operating and investment costs. Figure 5.1 depicts the technical aims of the beneficiation process. (Drzymala, 2007, p. 10 - 12)

Technical aims beneficiation:

reduce noxious substances out of the ore ↓

increase valuable substances in the concentrate ↑

meet requirements on the product concerning:

- equality
- content of valuable & noxious substances
- water content
- physical properties (particle size, particle form, surface area)

Figure 5.1 Overview technical aims beneficiation

Beneficiation of an ore includes the basic steps size reduction, classification separation and product conditioning. The ore is classified according to particle size and shape. The ore is separated according to gravity, solubility, discerptibility, mechanical, electrical, optical, surface or chemical properties. Table 2.6 lists example aggregates. (Drzymala, 2007, p. 10 - 12; Fuerstenau and Kenneth, 2003, p. 10)

Table 5.1 Overview separation property, method and example aggregate

Separation Property	Separation method	Example Aggregate
Discerptibility	washing / desilting	washing cylinder
Electrical	electro-static sorting	magnetic belt pulley
Gravity	sink-float process setting work sorting on tables and riffles	dense-medium separator jig washer sorting riffle
Optic	hand sorting sensor sorting	- x-ray transmission sorter
Solubility	leaching	heap leaching
Surface area	flotation	flotation cell

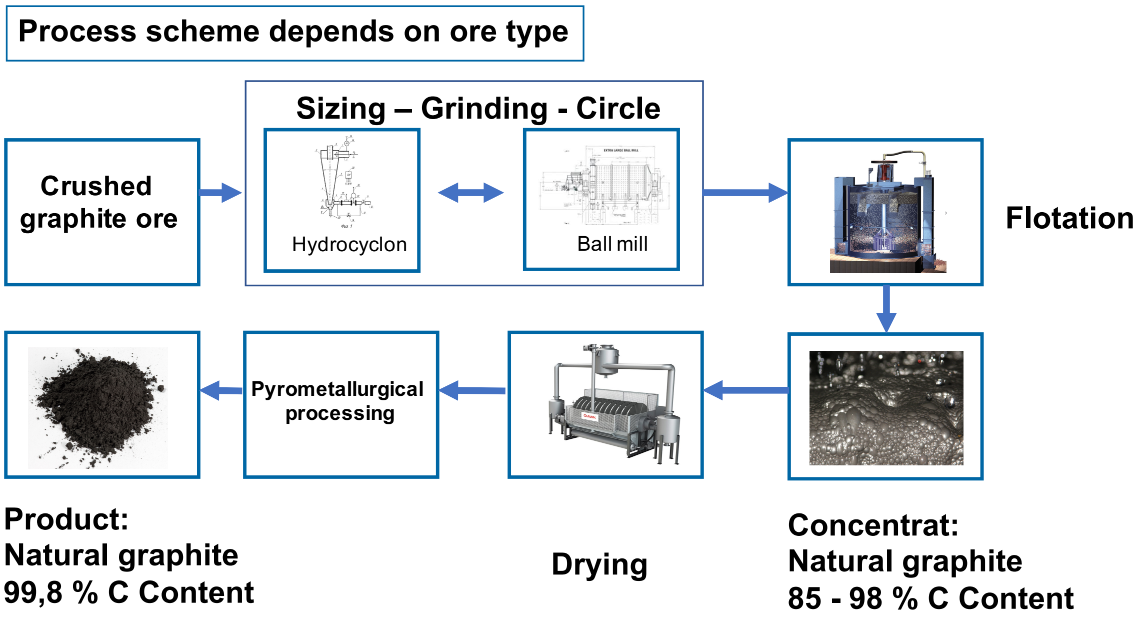


Figure 5.2 General process scheme natural graphite beneficiation (Fahl, 2017)

Figure 5.2 is a general schematic description of the graphite beneficiation process. The process depends mostly on the ore type and chemical properties of the natural graphite ore to be treated. The main six beneficiation process steps are crushing, sizing, grinding, froth flotation, dewatering and packaging. Carbon content of the flotation concentrate ranges from 85 to 98 % depending on the ore and process quality. Pyrometallurgical processing can be performed to increase the product's carbon content to 99,8 %. (Bulatovic, 2014, p. 164 - 169)

Crystalline flake ore is processed to create the most valuable marketable product. The flake size distribution within the ore is a debated factor. Scogings discussed the issue and explained the direct, linear relation between the flake size and the price of the marketable product as outlined in the Table 5.2. (Scogings b, 2017, p. 79)

Table 5.2 Examples of market prices linked to graphite product flake size and purity. Based on (Scogings b, 2017, p. 79)

Flake size in [µ]	Mesh Size	Purity	Price / tonne (US$)
>300	+48	90 to 97 %	~2,000
180 to 300	-48 to +80	90 to 97 %	~1,300
150 to 180	-80 to +100	90 to 97 %	~1,100
75 to 150	-100 to +200	90 to 97 %	~750
<75	-200	80 to 85 %	~450

5.1 Technical Process scheme: Beneficiation

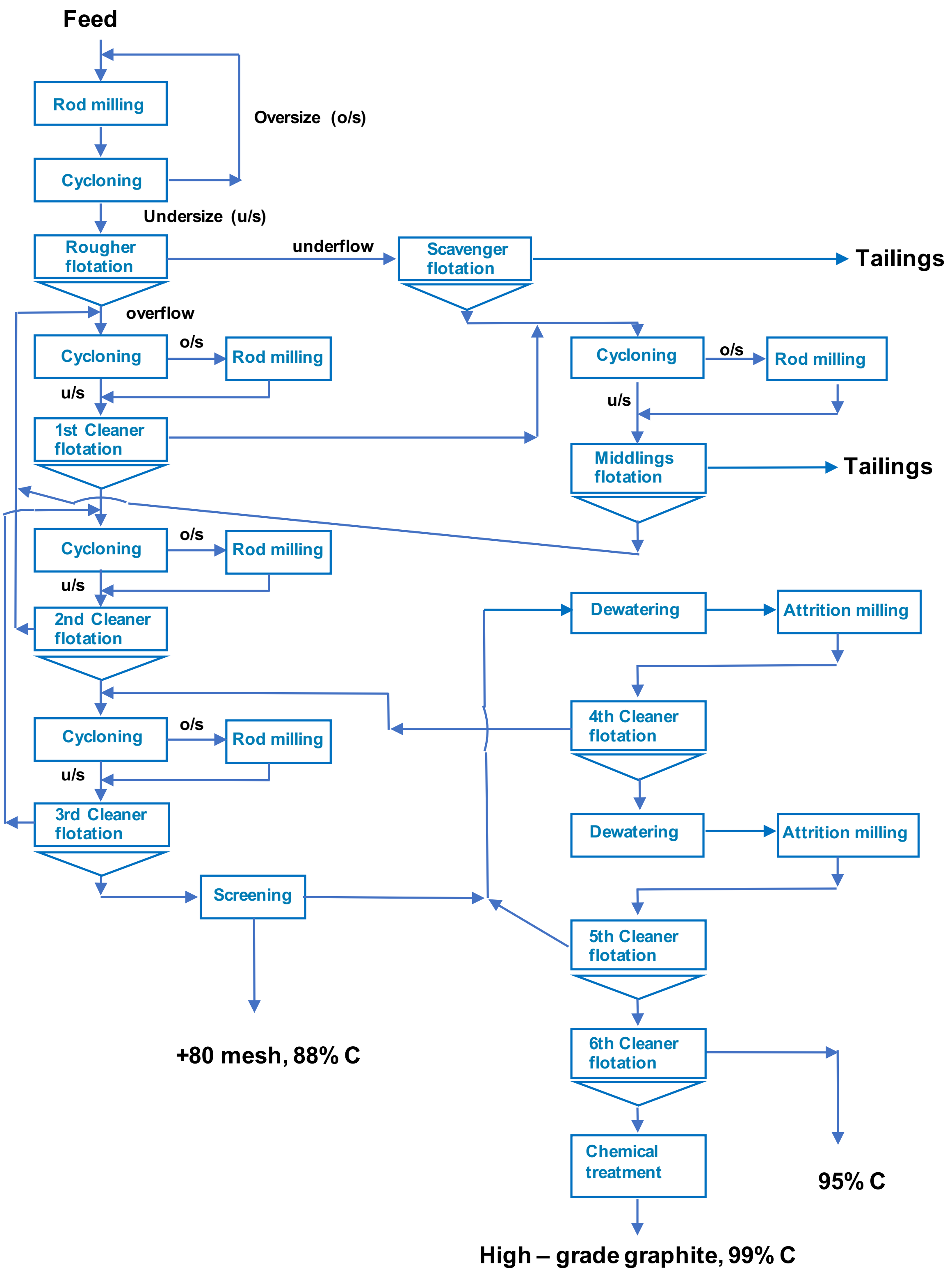


Figure 5.3 Detailed technical process scheme flake graphite beneficiation. Data from (Bulatovic, 2014, p. 168)

A detailed technical process flow sheet describing the process of coarse high-grade flake graphite beneficiation is Figure 5.3. Figure 5.4 outlines the counter current cascade structure and aims of the technical process of coarse high-grade flake beneficiation. The process includes ten flotation stages: one rougher, one middlings, six cleaner and one scavenger. Main aim of the process design is to prevent spreading of the soft natural graphite flakes inside the mills and to preserve the particle shape of the graphite flakes. Intact natural graphite flakes have a higher market price (see Table 5.2). Table 5.3 is a list of the process aggregates. (Bulatovic, 2014, p. 164)

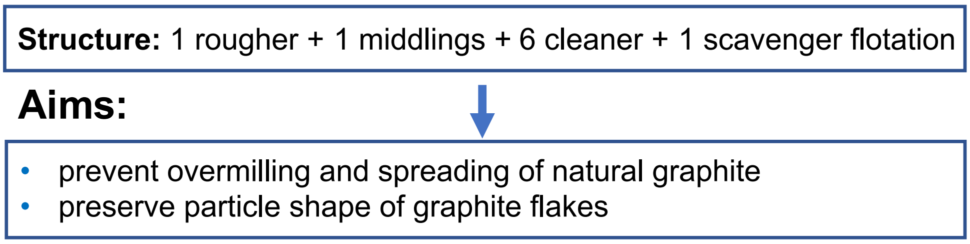


Figure 5.4 Structure and aim of the process of coarse high-grade graphite beneficiation

The process utilizes rod mills in the initial three flotation cleaning stages. In the last three cleaning stages attrition mills are used. Attrition mills pulverize the feed between toothed metal disks rotating in opposite directions. Attrition milling minimizes the spreading of graphite particles. Three types of coarse high-grade flake graphite are produced: a + 80 mesh (177 µm) 88 % carbon content concentrate, a concentrate assaying 95 % carbon content and a concentrate assaying 99 % carbon content after chemical treatment. (Bulatovic, 2014, p. 164; Chelgani et al., 2016, p. 60)

Table 5.3 Overview equipment process flow sheet. Based on data from (Bulatovic, 2014, p. 164)

Amount	Aggregate
5	Rod mills
2	Attrition mills
5	Hydrocyclone
9 Stages of	Flotation cells
1	Screen
Several	Filters
Several	Pumps
1	Reactor for chemical treatment

5.2 General process description

Worldwide about 100 natural graphite production plants operate. Different types and qualities of natural graphite products are produced. The product properties depend on the ore quality and the buyer's requirements. The process flow sheets vary. Figure 5.2, Figure 5.3 and Figure 5.6 show flow sheets of different complexity. The process feed's carbon content ranges from 3 % to 90 %. Through beneficiation a product with a carbon content of 98 % is produceable. The remaining 2 % impurities are intercalated metal vapor ions. Metal vapors intercalated between the graphite crystal layers can be separated by chemical or thermal purification methods. (Bulatovic, 2014, p. 166 - 167)

- **Reduction of particle size and classification**

The process route for amorphous/microcrystalline ore: Grizzly bars are used to separate bolders. The fine clay containing material is washed and grinder to a floatable size of under 1 cm. (Cirkel, 1907, p. 210)

The process route for crystalline flake natural graphite: The precrushing process depends on the ore type and mining method. Several slow speed regrind mills are used to prevent overgrinding. Slow speed regrind mills are attrition mills or ball mills equipped with pebble stones. (see Chapter 5.1) (Bulatovic, 2014, p. 165)

The process route for vein-type natural graphite ore: The ore is crushed under brittle stress. The extracted lumps are hand-cobbed on a sorting patio to remove quartz inclusions. In mechanized operations probably jaw crushers are used. The compact construction of jaw crushers enables the underground usage. An alternative is the usage of impact crushers. Due to the low flow rates the use of a gyratory crusher is not useful. Semi-autogenous mills are used. Compared to rod mills semi-autogenous mills have a better energy balance. * (Taylor, 2006, p. 514)

*[Personal comment: No specific mechanized process is described in literature. The described process route is an assumption made considering the mohs hardness of the ore and the mining conditions [Fahl]]

- **Froth flotation**

Liberated crystalline natural graphite flakes are hydrophobic and naturally floatable using frothers. No collector is used. The enclosing silicate rock is hydrophil. Silicate rock is depressed with sodium silicate or lactic acide. Enclosing calcite rock is depressed with lignin sulfonite. Table 5.4 lists the reagents used in froth flotation. Froth flotation is performed in an alkaline environment (pH value 7,5 - 8,5). The optimal particle size range for a successful froth flotation of a low density material is 1.600 µm to approximately 35 µm. The minimal theoretical particle size is 1 µm. (Bulatovic, 2014, p. 164; Chelgani et. al, 2016, p. 60 - 62; Wang et al., 2010, p. 70; Drzymala, 2007, p. 348)

Table 5.4 Froth flotation reagents. Based on data from (Bulatovic, 2014, p. 164)

Reagent	Function
MIBC, pine oil, fuel oil, kerosene	frother*
Sodium silicate, lactic acide, lignin sulfonite	depressant*
None	collector*

Figure 5.5 depicts the construction and function of a flotation cell. An air bubble stream is produced. The hydrophobic crystalline natural graphite flakes form Van der Waals forces with the air bubbles. The air bubbles carry the liberated hydrophobic natural graphite flakes to the surface of the flotation cell. The natural graphite loaded froth, the flotation concentrate is paddled out of the flotation cell. (Chelgani et. al, 2016, p. 60 - 62; Schubert, 1996, p. 379)

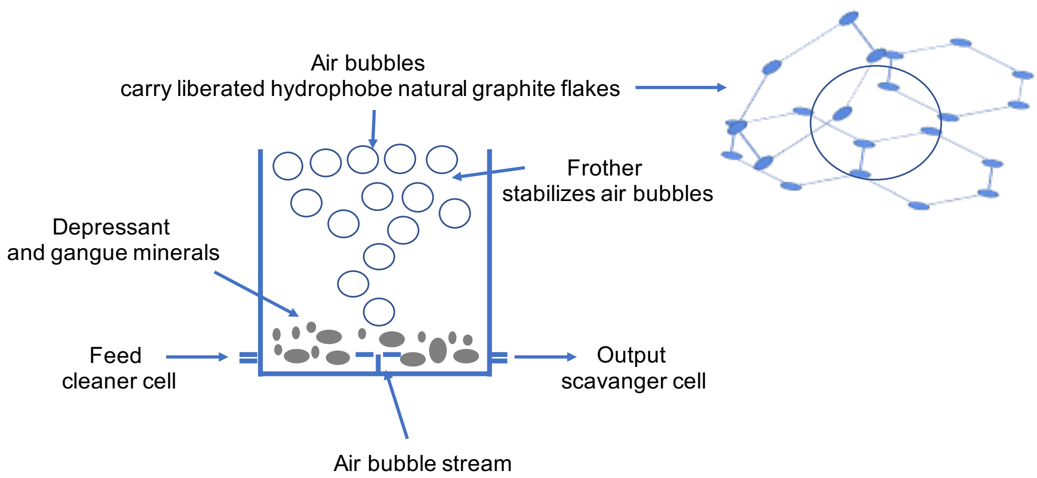


Figure 5.5 Schematic model flotation cell. Based on data from (Schubert, 1996, p. 379)

- **Dewatering & Packaging**

The flotation concentrate is filtered. Table 5.5 lists examples of filters. The process water is recycled. The transportation ability of the flotation concentrate is increased. Transportation costs are reduced. **Thermal drying** is necessary to archive a water content under 15 %. Thermal drying is energy intensive. **Thickener** with flocking substances can be used to save energy costs. Requirements on the product's water content depend on the transport and trade routes and the purification method. Industrial minerals are normally traded in 30 kg Bags or 1013 kg Big Bags. (Kogel et al., 2006, p. 99; Taylor, 2006, p. 516)

Table 5.5 Filter aggregates with personal evaluation

Filter aggregate	Advantages	Disadvantages
Vaccume membrane filter	High flow rate, continuous process, compact construction	High investment costs
Camber press filter	Low investment and operating costs	Old technology, batch process
Belt filter press	Very high flow rate, integrable in continuous process	High investment costs

- **Possible alternatives to the conventional process**

Dense media separators are the most efficient industrial gravity-based separators used for coal cleaning. Graphite, which has similar properties as coal, is floatable in Dense media separators. The use of Dense media separators is a possible alternative for the beneficiation of low-density graphite ores with less gangue minerals and higher carbon content. Additional research is still required. (Chelgani et. al, 2016, p. 64)

Tran et al. 2010 conducted a study to demonstrate that it is possible to selectively separate ultra-fine graphite with a method called **film flotation**. Graphite particles cluster at the water surface. Ultra-fine particles can be recovered at the saturated water surface. (Chelgani et. al, 2016, p. 64)

- **Purification and further treatment**

To create a graphite product with a carbon content of 98 - 99.8 % from crystalline flake flotation concentrate further purification is necessary. The purification methods separate the intercalated metal vapors (e.g. lithium, natrium, sulfur) from the crystal lattice. (Chelgani et. al, 2016, p. 62 - 63)

The wet **chemical leaching** with hydrochloric (HCl) and hydrofluoric (HF) acid is the most common purification technique to produce high-purity graphite from flotation concentrates. Hydrofluoric acid removes silicates and is then washed and filtered to achieve the graphite separation. Hydrochloric acid converts the metal impurities in soluble chloride. Additional alkali **roasting** is required to remove sulfidic impurities. Flotation concentrate from Woxna Graphite AB was tested by Lu et al. to find the ideal process parameters. The ideal parameters found out are: roasting with NaOH at 250 °C followed by water washing and H_2SO_4 leaching. The sulfur content was reduced to 0.05 %. The already high-purity of vein-type natural graphite ore (95 - 99 % carbon content) can be further enhanced by alkali roasting. (Chelgani et al, 2016, p. 62 - 63; Lu et al, 2002, p. 757; Balasooriya et al., 2015, p. 163; Wei Xie, 2015, p. 53)

The chemical leaching and roasting have inexpensive operating costs due to the low energy consumption. Both processes cause serious environmental damage (see Chapter 6.1). (Chelgani et al, 2016, p. 62 - 63)

Four **thermal purification** methods are commonly used: a variant of the **acheson process**, the **fluidized bed process** and the **thermal halogenation and chlorination gasification processes**. All thermal purification methods have expensive operating costs due to the high energy consumption. Thermal purification methods cause less environmental damage.

During the **Acheson process** the natural graphite flotation product is pelletized. Fine graphite particles increase the danger of combustion. The graphite pellets are mixed with coke. The mixed material is placed between two electrodes and covered by refractory powder to prevent oxidation. Electric current is conducted through the mixture. The mixture acts as electric resistance. Temperatures above 3,000 °C are generated. The impurities react with the coke during the graphitization process. Purities of 99.9 % carbon content are achieved. (Ambrosi et al., 2012, p.1)

During the high-temperature **fluidized bed process**, the natural graphite flotation product is heated up to 2400 - 2600 °C under inert gas or vacuum atmosphere. The temperature of 2400 - 2600 °C is over the boiling point of the intercalated alkali metals. The intercalated alkali metals vapor out of the product. (Fedorov, 2014, p. 12)

Gasification is a favorable process for graphite products with high fine grain share. The high surface area is advantageous for the interfacial diffusion of the gaseous reagent or volatile liquid. During the low-temperature **thermal halogenation** at temperatures above 500 °C gaseous halogens or halogen compounds are added to a natural graphite flotation product in a reactor. The intercalated impurities are transferred into volatile halogen compounds (halides) and removed. During the low-temperature **thermal chlorination,** a natural graphite flotation product is heated in the presence of chlorinated hydrocarbons having a boiling point above 200 °C. At a temperature of at least 1000 °C chlorides are formed from the impurities and volatilized. (US 3035901 A, 1962; Ambrosi et al., 2012, p. 5)

5.3 Case study: Nanshu Mine

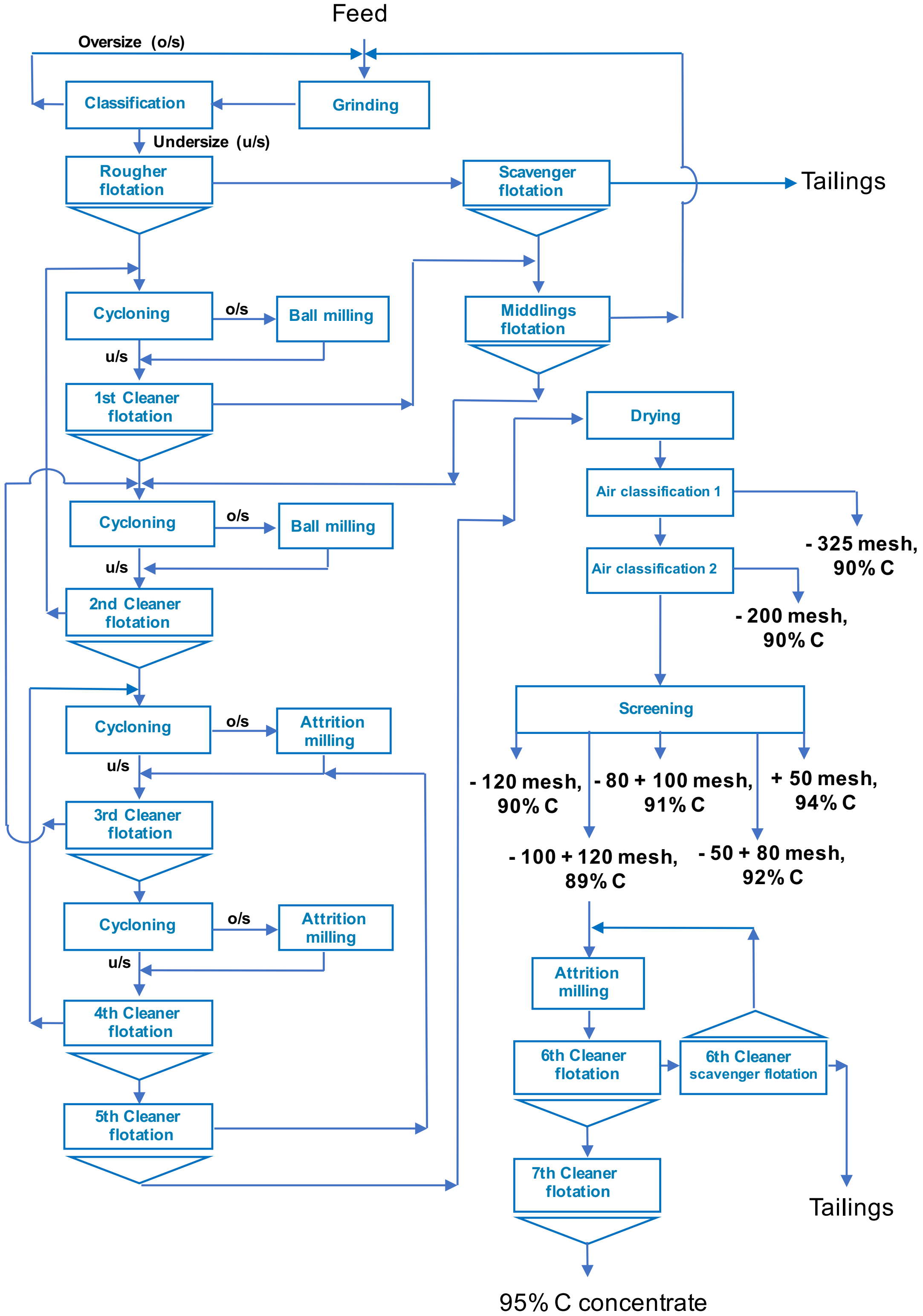


Figure 5.6 Detailed technical process flow scheme Nanshu Mine. Based on (Bulatovic, 2014, p. 168)

Table 5.6 Data sheet "Nanshu Mine". Based on (Hudson Institute of Mineralogy, 2015, p.1)

Nanshu Surface Mine

Geographical location: 36°57' N,120°22' E

Shandong Province, China

Figure 5.7 Satellite picture "Nanshu Mine" (Google d, 2017)

Figure 5.8 Map section geographical location "Nanshu Mine" (Google e, 2017)

Local geology:

Paleoproterozoic Jingshan Group consisting of:

- marble and amphibole-plagioclase gneiss intercalated with graphite gneiss
- amphibole-plagioclase intercalated with marble and graphite gneiss
- marble and amphibole-plagioclase gneiss

Origin: Metamorphism of organic carbon, in clastic sedimentary rock, deposited in a shallow marine environment.

Host rock: Marble and gneiss

Minerals: Gneissic banded and granoblastic structured.

Ore body layout: 1000 m long bedded and multiply layered crystalline flake graphite masses, down dip extends from 50 to 400 m

Table 5.6 gives an overview of the location, the geologic features and layout of the ore body of the “Nanshu” Surface Mine. The “Nanshu” Surface Mine is located in the Shandong Province in northwestern China (see Figure 5.7 and Figure 5.8). From the bedded and multiply layered ore body amorphous and crystalline flake graphite ore is extracted. Figure 5.6 is a detailed technical process flow sheet describing the production of seven different crystalline flake flotation concentrates. The process includes ten flotation stages: one rougher, six cleaner and one scavenger. After the 5th cleaner flotation stage, intermediate drying, air classification and screening is used to separate the flake sizes listed in Table 5.7. (Hudson Institute of Mineralogy, 2015, p.1; Bulatovic, 2014, p. 168)

Table 5.7 Flake sizes: crystalline flake flotation concentrates Nanshu Mine*. Data source (Bulatovic, 2014, p. 168)

Flake size in [µ]	Mesh Size	Purity	Separation Aggregate	Market Term
> 300	+50	94 %	Screening	Coarse Flake
180 to 300	-50 to +80	92 %	Screening	Large Flake
150 to 180	-80 to +100	91 %	Screening	Medium Flake
75 to 150	-100 to +200	89 %	Screening	Small Flake
< 125	-120	90 %	Screening	
< 75	-200	90 %	Air Classification	Fine Flake
< 44	-325	90 %	Air Classification	

*[Personal comment: European sieves are standardized in micrometer. The standards are ISO 565:1990, ISO 3310-1:2000 (International) and EN 933-1 (European). American sieves are standardized in mesh (ASTM E11:01). [Fahl]]

6 Sustainability - Environmental aspects related to mining and beneficiation

In 1987 the Brundtland report, “Our common Future’’, laid the groundwork for our current definition of sustainability. The need of sustainable development defined as the notation:

> […] development that meets the needs of the present without compromising the ability of future generations to meet their own needs (Brundtland Commission, 1987, p. 254).

implies challenges and conflicts for the international mineral resource supply and the mining industry. Figure 6.1 symbolizes the balance of the three aspects of Sustainability: Environment, Society and Economy.

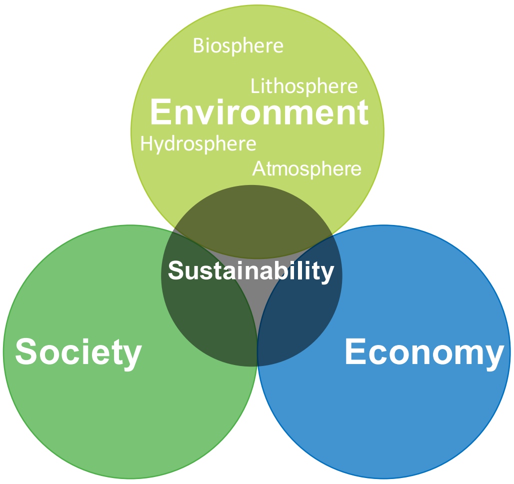


Figure 6.1 Three balanced spheres of sustainability

Challenges and Conflicts related to mining activities and sustainability cover serval topics: e.g. the influence on the global ecosystem and biodiversity, the environmental geochemistry and “how to get a Social License to operate’’. Challenges occurring in the long-term mine planning and daily operation are: e.g. the reduction of energy and fuel consumption and the organization of a responsible mine waste handling. Reclamation is a concern involving topics of all three spheres of sustainability. (Bell et al., 2006, p. 355; Kesler, 2015, p. 54; Ripley, 1996, p. 47, p. 110 - 111)

Mining companies commit to voluntary sustainability commitments and aims like fashion companies. Mining companies publish yearly sustainability reports to promote their progress. Sustainability progress in mining is measured with indicators: e.g. the animal population in an conserving habitat. (Global Reporting, 2011, p. 8)

6.1 Description of environmental aspects in general

The environmental challenge in graphite mining is to reduce the impact of the mining activities on the **atmosphere, biosphere, lithosphere and hydrosphere**. Emissions into the atmosphere include for example noise emissions from mining equipment or noxious graphitic fine dust emissions from uncovered waste piles. Visible impact on the lithosphere is the radical change of the landscape. Artificial landscape buildings for example point cone waste and stockpiles radically change the original landscape layout. Preserving a satisfying water quality and quantity is essential to reduce the impact on the hydrosphere. Sufficient filtering of the process water from beneficiation, acid leaching and alkali roasting is essential. The conservation of endangered species is essential to reduce the impact on the biosphere. The constant monitoring and controlling of environmental indicators (e.g the population of animals in an conserving habitat) is necessary to run a responsible and sustainable mining operation. (Bell et al., 2006, p. 355; Kesler, 2015, p. 54; Ripley, 1996, p. 47, p. 110 - 111)

In practice a major environmental problem during graphite mining is the waste material handling. The feasibility study of the “Lac Knife exploration’’ project estimates a weight recovery of 13,7 % during the mining and beneficiation progress. The remaining 86,3 % weight percentage must be disposed as tailings. Problematic is the disposal of fine tailings. Fine tailings must be sedimented in tailings ponds to assure an appropriate handling of toxic ingredients. Coarse agglomerates can be used for construction works to reduce the necessary tailings pond capacity. (Desautels et al., 2014, p. 169; Tripathi et al., 2016, p. 27 - 28)

The riverine disposal of tailings is common practice in the Chinese graphite mining industry. Mine water disposed in rivers can cause undesirable turbidity, undesirable sedimentation and toxic effects on vegetation and animals. (Tripathi et al., 2016, p. 27 - 28; IMO, 2013, p. 25)

Intense energy consumption during the beneficiation and processing of natural graphite ore is another concern: The total energy amount required to produce 1 tonne of natural graphite crystalline flake product (approximately 99,5 % carbon content) is 2500 kwh/t. Remotely located mining operations are commonly supplied with energy from diesel engines. Energy supply from renewable energy sources (except hydropower) is uncommon. (AMG, 2015, p.16)

6.2 Economic and social challenges of the graphite mining industry

Social challenges the graphite mining industry needs to resolve are for example:

- the acceptance of individual cultural, legal and normative conventions (depending on the graphite mining country)
- allowing the participation of the local population (e.g. by allowing influence on decisions and offering labour)
- maintain transparency about the mine plan, mining methods and used technologies

The graphite mining industry in China is an interesting example to evaluate economic and social challenges of the worldwide graphite mining industry**.** China has a monopolistic market position with a market share of 69 % in the volatil natural graphite market. Most important graphite mining regions in China are the Heilongjiang and the Liu-Mao provinces. Approximately 200 companies produce natural graphite in China. About 10 major Chinese natural graphite exporters operate globally. Due to environmental pollution the Chinese government closed 55 amorphous natural graphite mining operations in 2013. The 55 mining operations contributed a share of about 10 % to the worldwide natural graphite production. The Chinese government plans to restore the fragmented graphite mining industry to support the economic growth. Driven by the economic growth mining companies need to solve the trade off between the rising demand for higher wages and cost-efficient production. The Chinese government introduced an export duty of 20 % on natural graphite product exports to prevent the early depletion of the Chinese natural graphite reserves. (USGS a, 2017, p. 75; Olson, 2016, p. 71)

[*of the worldwide natural graphite production in 2014 (USGS, 2017 a, p. 75)]

6.3 Jongchon Mine - Analysis of satellite photographs between 2006 and today

Criteria for an environmental evaluation of satellite pictures are inter alia the analysis of any:

- visual impacts on vegetation
- visual signs of water pollution
- visual signs of instability of constructions (e.g. tailings ponds)

Figure 6.2 is a timeline of aerial satellite pictures of the "Jongchon Mine". The timeline depicts the development of the mining facility and the tailings pond. Figure 6.3 is a 3D satellite picture of the "Jongchon Mine" and landscape. (see Appendix F for high resolution satellite photographs)

Challenging for the water management of the "Jongchon Mine" is the short and intense precipitation period during the southern monsoon in summer. Table 6.1 is an evaluation of the environmental situation visible on the satellite pictures. (Lee et al., 2017, p. 2)

Table 6.1 Evaluation environmental situation "Jongchon Mine" 2006 - 2017

2006	Start of the development of the mine. The process plant and transformation station connected to the Pyongyang - Yonan electricity link is constructed. Overburden is pre-stripped north of the process plant for the construction of a tailings pond.
2009	The area surrounding the mine is covered with vegetation. Four ponds with highly sediment loaded waste water are constructed next to the process plant. Mining activities are conducted south of the process plant.
2010	No significant mining progress
2012	No significant mining progress
2013	Stable walls of the raised embankment tailings pond are visible. The mining facility is surrounded by agricultural vegetation. Trees are planted next to the process plant. The four waste water ponds next to the process plant are recultivated and vegetated. The stripped area with visible active mining is south of the processing plant.
2017	The "Jongchon mine" is fairly embedded into the landscape and fully surrounded by agricultural vegetation. No destructive, inappropriate constructions like cone waste piles are visible.

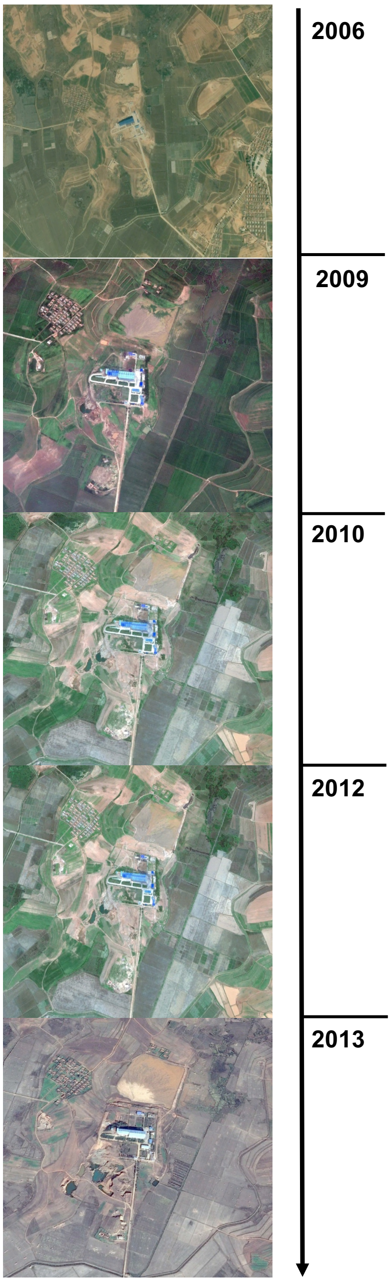


Figure 6.2 Timeline: Aerial satellite pictures “Jongchon Mine” from 2006 to 2013

Figure 6.3 Aerial satellite picture “Jongchon Mine“ and landscape 2017

7 Evaluation of the current criticality of the European natural graphite supply

Natural graphite is defined as a critical raw material. The European Ad-Hoc Working Group on defining critical raw materials has listed twenty critical raw materials. Figure 7.1 is a diagram sorting the selected critical raw materials. The European Ad-Hoc Working group applied two criteria: the economic importance and the supply risk of the selected raw materials. (Ad hoc Working Group, 2014, p. 3 - 4)

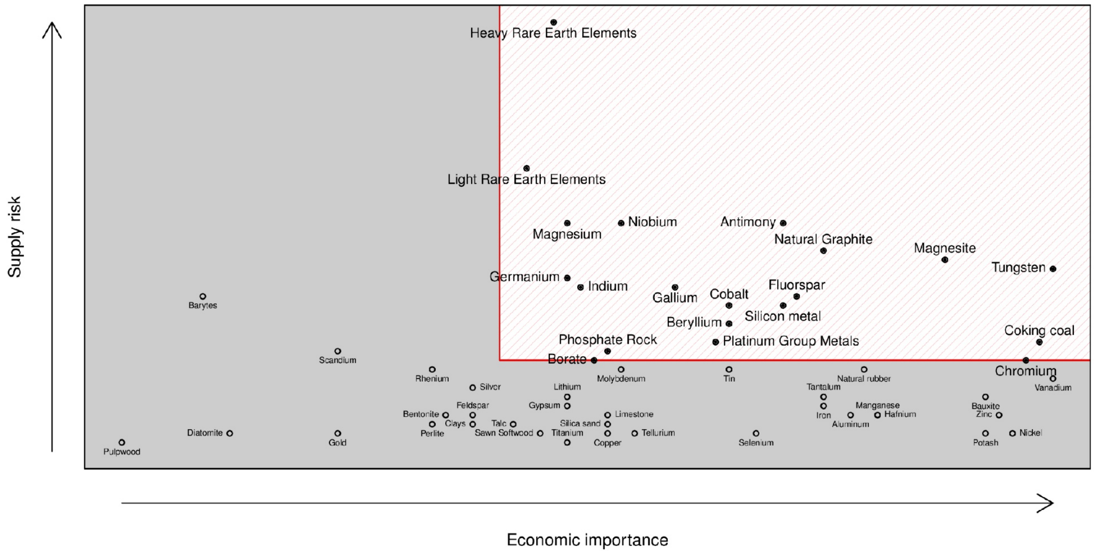


Figure 7.1 Diagram: European critical raw materials (Ad hoc Working Group, 2014, p. 3 - 4)

The static lifetime of natural graphite reserves can be calculated by the division of the amount of reserves through the annual production rate (Figure 7.2). Natural graphite is a geologically fairly abundant mineral. The natural graphite supply has growth potential. New exploration projects and mines are developed worldwide, especially in Canada and Africa. The criticality of the European natural graphite supply mainly depends on the volatil market situation and the Chinese exports. European post-processors secure their natural graphite supply with buyer-seller contracts.*

Static lifetime of natural graphite reserves:

76.800.000 t ÷ 1.130.000 t/a = 68 a

Figure 7.2 Equation static lifetime of natural graphite reserves in 2014. Data from (USGS a, 2017, p. 75)

[*Personal evaluation. [Fahl]]

7.1 Current challenges of the graphite mining industry - general outlook

The worldwide natural graphite consumption steadily increased from 2012. The reason for the increase is the improvement of global economic conditions. Figure 7.3 illustrates the linkage between the worldwide steel demand and the worldwide natural graphite demand. A share of 58 % of the global natural graphite demand in 2012 was used in steel applications. Figure 7.4 gives an overview of the volatile market prices in the period from 1995 to 2015. The U.S. Geological Survey did not release market prices for 2016 and 2017. (Shaw, 2015, p. 6; USGS c, 2017)

In 2012 a share of 4 % of the global natural graphite demand was used for the production of anodes for different battery types. A share of 80 % of the natural graphite used in batteries in 2012 was utilized in lithium-ion batteries. The automotive company Tesla plans to produce 500,000 electric vehicles by 2050 in a manufacturing plant in Nevada. An amount of 30 - 60 kg natural graphite is installed in a lithium-ion electric vehicle battery. Compared to the metals utilized as cathode material natural graphite is an inexpensive component. According to the U.S. Geological Survey the production will require 93,000 tonnes of flake graphite to produce anodes for lithium-ion batteries. (Tesla, 2017; USGS a, 2017, p. 75; Rothermel et al., 2016, p. 3481)

Apart from the use in lithium-ion batteries possible new high technology applications for high purity graphite powders could led to a growing graphite demand. Likely is the use of large quantities of natural graphite in fuel cell applications, carbon-graphite composites, electronics or speciality lubricant applications. (USGS a, 2017, p. 75)

Recycling techniques for lithium-ion batteries are investigated. For example during the European research project LithoRec 2. Rothermel et al. analyzed the technical recycling process. The production of recycled graphite from lithium-ion batteries is technically possible. The future discussion is wether the recycling of lithium-ion batteries is more reasonable and cost efficient than their disposal. (Rothermel et al., 2016, p. 3481)

7.2 Overview of market prices 1995 - today

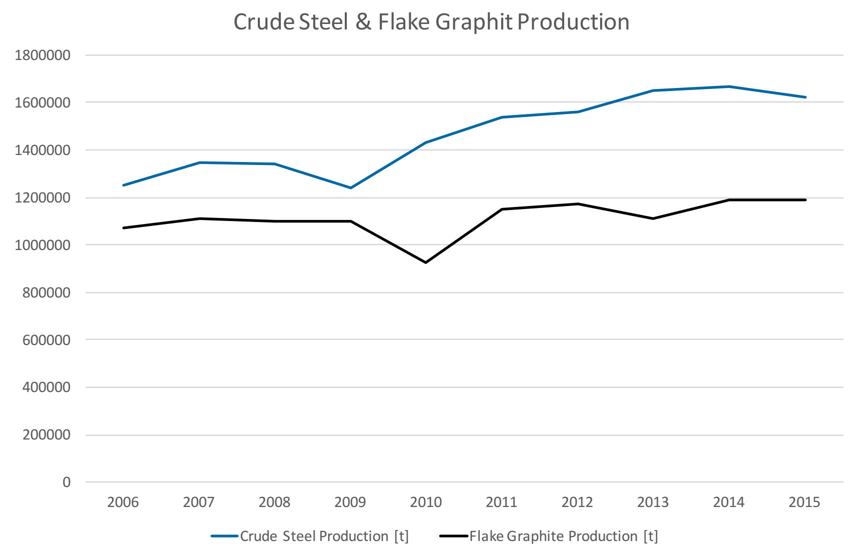


Figure 7.3 Diagram Crude Steel Production 2006 - 2015. Based on data from (USGS c, 2017; World Steel Association, 2016, p. 2)

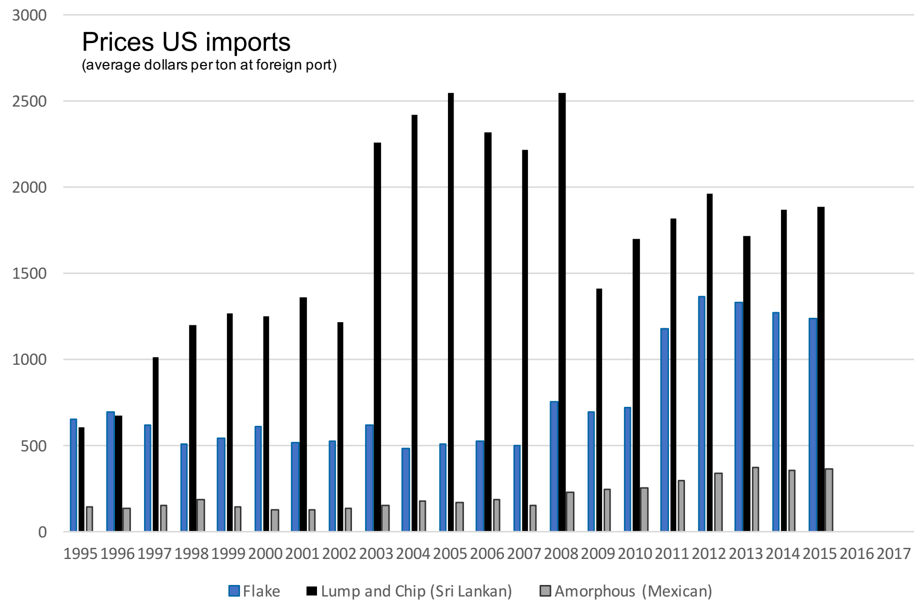


Figure 7.4 Diagram US import prices natural graphite. Data from (USGS c, 2017)

8 Conclusion

Natural graphite is an allotrope of pure carbon that crystallizes in the hexagonal crystal lattice, with rhombohedral symmetry. Natural graphite is defined as a critical raw material by the European Union and has a high HHI-Index of 5.015. Natural graphite is commonly categorized by particle size in the three categories: crystalline flake, amorphous and vein-type.

8.1 General conclusion

Mineral occurrences are geologic anomalies. Natural graphite occurrences and deposits are formed by the maturation and metamorphism of immature organic material and precipitation from C-O-H fluids. Vein-type natural graphite deposits are found in metasedimentary belts, crystalline flake deposits are found in paragneiss and marble, that have been metamorphosed to amphibolite and granulite facies. Amorphous graphite particles are distributed in weakly metamorphosed rocks. Most European natural graphite occurrences and deposits are found in the northern countries. The bedrock is part of the “Fennoscandian Shield’’ a stable rock area consisting of Precambrian rocks.

Natural graphite is extracted in underground or surface mines. The mining method depends on the locational, natural and geologic factors. Geologic factors include criteria such as topography, spatial relations of the ore body, mineralogy, petrography and rock mechanics properties. Natural graphite mines are small scale or artisanal mining operations. The extraction of natural graphite includes the four process steps excavation, transporting and primary crushing. The beneficiation of natural graphite includes the six process steps crushing, sizing, grinding, froth flotation, dewatering and packaging. Through beneficiation a product with a carbon content of 98 % is produceable. To create a graphite product with a carbon content of 98 - 99.8 % thermal and chemical purification methods are used.

A major environmental problem concerning the graphite mining industry is the waste material handling. The riverine disposal of tailings is common practice in the Chinese graphite mining industry. Mine water disposed in rivers can cause undesirable sedimentation and toxic effects on the biosphere. The international natural graphite market is volatil and the demand for natural graphite is likely to grow. China has a monopolistic market position.

8.2 Potential for additional research

The further geological evaluation of European natural graphite deposits and occurrences is still an interesting and important research objective. The production and collection of thin sections from European graphite ores to determine the economically important flake size distribution is an important step. Conducting intensive exploration campaigns including beneficiation and metallurgy tests is the next step. The development of enhanced froth flotation methods, energy efficient purification methods and economically feasible recycling methods for lithium-ion batteries offers enormous research potential.

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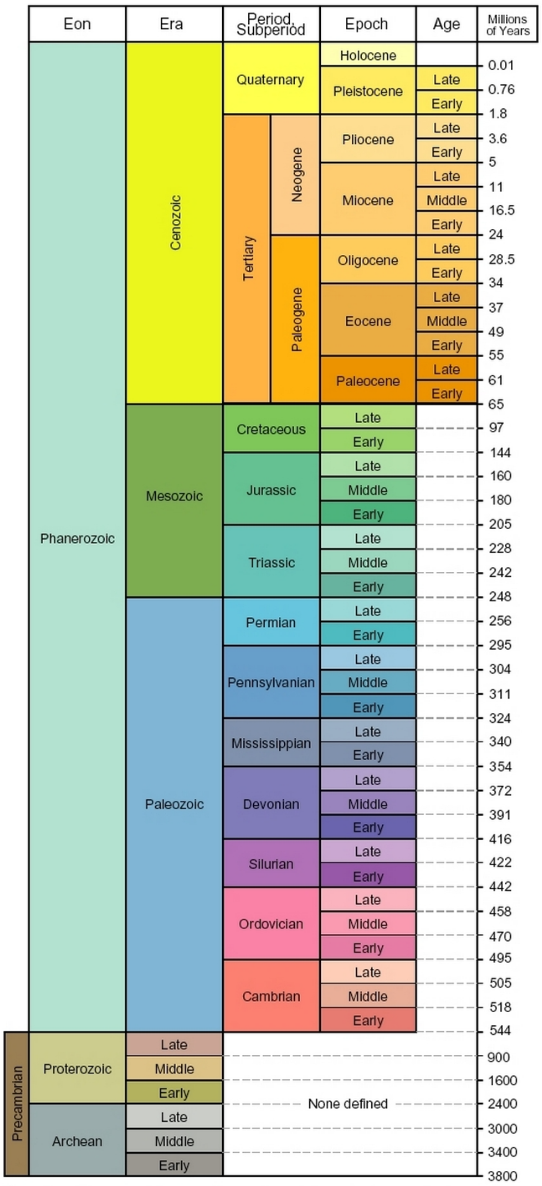
Appendices

Appendix A: Definition HHI-Index (U.S. Department of Justice, 2015)

The term “HHI'' means Herfindahl-Hirschmann Index, a commonly accepted measure of market concentration. The HHI is calculated by squaring the market share of a country on the global resource market and adding the resulting numbers. For example on a resource market with four dominating countries with shares of 30, 30, 20 and 20 percent the HHI is 2.600 ($30^2+30^2+20^2+20^2 = 2.600$)

Appendix B: Diagram geotimes (Hakki, 2012)



Appendix C: Overview prospection and exploration reports Woxna deposit

(Reed, 2015, p. 117 - 118)

	Year	Report name	Author*	Company
Prospection	1983	-	-	-
Exploration	1988	Drilling & Exploration Report Kringelgruvan	Claesson	-
	1989	Drilling & Exploration Report Kringelgruvan	Claesson	-
	1989	Drilling & Exploration Report Kringelgruvan	Claesson	
	1991	Drilling & Resource estimate report Gropabo	Claesson	
	1992	Drilling & Resource estimate report Mänsberg	Claesson	
	1993	Drilling & Resource estimate report Mänsberg	Claesson	
	2001	Feasibility Study, Graphite Leaching	Boliden Contech	Boliden Contech
	2001	Experimental study on the upgrading of fine Woxna concentrate	Lu	-
	2002	Technical Study, Graphite Leaching	Boliden	Boliden
	2002	Woxna Graphite Mineral Ressources Report	Claesson	-
	2011	Technical Report Woxna Graphite Project	-	Coffey Mining Pty Ltd.

	2011	Woxna Graphite Project Report on Geology, Mineralization and Exploration Potential	Thamm	Coffey Mining Pty Ltd.
	2012	Technical Report: Kringelgruvan	Reed	
	2013	Technical Report Woxna Graphite	-	GBM Minerals Engineering Consultants
	2015	Technical Report Woxna Graphite	Reed	Reed Consulting

* only one name given

Appendix D: Glossary

(Lovering, 1963, p. 1; West, 1982, p. 1-10; Schlumberger, 2017; Schubert, 1996, p. 240)

Term	Definition
Carbonization	**Chemical explanation:** The formation of carbon from organic substances through pyrolysis or destructive distillation. **Geology**: Term equals Maturation process
Collector	Reagent used during Flotation to increase the hydrophobicity of the surface area of the valuable substance.
Depressant	Reagent used during Flotation to increase the hydrophilicity of the surface area of the gangue minerals.
Frother	Reagent used during Flotation to form air bubbles transporting liberated natural graphite particles.
Graphitization	**Chemical explanation:** The formation of crystallized C-H rings.
Maturation	The process of carbon hydroxide generation from a host rock under pressure and temperature of the burial.
Metamorphism	The process that changes the features of a rock or recrystallizes it.
Precipitation	The formation of an insoluble material in a fluid. Precipitation occurs by a chemical reaction of two ore more ions in a solution or by changing the temperate of a saturated solution.
Syngenetic formation	The mineral crystallizes at the same time as the host rock. The mineral crystallizes during lithification.
Ternary plot	A ternary plot is a barycentric plot on three variables summing to a constant. The ratio of the three variables is depicted.

Appendix E: Definition of Units

All Units are according to the metric SI-System.

Unit (abbreviation)	**Explanation**
Tonne (t)	A metric tonne is a weight measure containing 1.000 kg.
ROM Ore	The Term "Run of Mine Ore" means Ore of a size that can be sold or further processed without additional crushing.
Dry t per year	Metric tonnes of dried material produced annually

Appendix F: Satellite photographs "Jongchon Mine"

Aerial Satellite Picture 2007

Aerial Satellite Picture 2009

Aerial Satellite Picture 2010

Aerial Satellite Picture 2012

Aerial Satellite Picture 2013

Aerial Satellite Picture 2017