Chapter 11 The Evolutionary Types of Magmatic Complexes and Experimental Modeling Differentiation Trends



N. I. Bezmen and P. N. Gorbachev

Abstract Many minerals are associated with massifs of magmatic origin, in particular, sulfide copper-nickel ores, chromite and titanium-magnetite deposits, deposits of the platinum group elements (PGE), etc., so many aspects of petrology, in particular petrochemical analysis of the structure and differentiation of massifs are also gaining economic importance in addition to petrological. Despite the great success in the study of the magmatic formations of specific regions: the Baltic Crystal Shield, the Voronezh Crystalline Massif, Transbaikalia and Siberia, South Africa and other regions of the world, many geological, petrological and geochemical features of the evolution of magmatic massifs have not yet been fully studied, or performed at a low physical and chemical level. In the present article the petrochemical systematization of intrusive magmatic complexes has been developed, which became the basis for the petrologic and metallogenic classification of differentiated intrusions with the separation of massifs, promising for the content of ore mineralization.

Keywords Experiment \cdot Intrusions \cdot Petrology \cdot Diagram \cdot Norilsk \cdot Stratified intrusions \cdot Differentiation

11.1 Introduction

Petrochemical systematization of massifs was based on the types of intrachamber differentiation, which are clearly revealed on petrochemical diagrams by trends in the evolution of magmatic complexes of different formations. As it was shown experimentally, superliquidus nanocluster differentiation of fluid saturated melts occurs at the level of complexes (associates, cybotaxises) close to minerals in structure (Bezmen 1992; Bezmen and Gorbachev 2017). The existing petrochemical diagrams, for which a limited number of components are used, are not quite suitable for studying the processes of intrachamber differentiation of melts, as they are usually used to

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generalize the normative compositions of rock-forming minerals: olivine and pyroxene on the one hand, feldspartoids and quartz on the other. In this connection, we have developed a petrochemical diagram on which almost all normative rock-forming minerals (olivine, pyroxene, plagioclases, quartz, nepheline, leucite, spinel, chromite, titanomagnetite, etc.) are fixed by figurative points.

11.2 Methodological Features

11.2.1 Petrochemical Diagrams Construction Method

The whole spectrum of stratified rocks of the massifs (ultrabasic, basic, acidic, alkaline and even ores) is compared on the diagram. It is based on the separation of femical components, sodium-calcium feldspar and silica. Since the role of calcium is ambiguous: on the one hand, it is concentrated together with aluminum, bind in the main plagioclase, on the other—is accumulated together with iron in clinopyroxenes, the total calcium content was divided by calculation:

$$Ca^{P1} = 0.5(Al + Na + K), \quad Ca^{Px} = Ca - 0.5(Al + Na + K).$$

On the diagram $Mg + Fe + Mn + Ca^{Px} + Cr + Ti + P(Na + K + Ca^{P1} + Al) - Si$, in at.%, petrochemical differences of rocks are clearly distinguished, as all main rock-forming elements are used.

The some interesting trends of superliquidus nanocluster differentiation were confirmed experimentally.

11.2.2 Methodology of Experiment

As a rule, the fluid phase of massifs contains water, hydrogen, fluorine and other fluid components, so the experiments were conducted in the installation of high gas pressure at the controlled fugacities of gases in the system H–O–C–F–Cl, the main components of the magmatogenic fluid. The peculiarity of the experimental method was the direct dosing of hydrogen in the composition of the fluid by means of its diffusion through the walls of platinum ampoules with the use of improved methods of the Show membrane (Bezmen et al. 2016) and the control of fugacities of other gases in the system H–O–C buffer reactions with carbon or Ni–Fe in the presence of hydrogen-containing visitie. The scheme of a hydrogen cell with a charged ampoule is described in detail in the works (Bezmen 2001; Bezmen et al. 2016). The glass powder made of oxides, corresponding to the weighted average composition of the trend rocks, was used as the initial silicate composition. In order to avoid iron diffusion into the platinum ampoule from the melt during the experiment, the experiments

were carried out in vitreous carbon or Ni–Fe of the alloy crucibles, which were also indicators of carbon activity or oxygen fugacity. Fluorine and chlorine were injected as an acid solution. The crucible was inserted into a platinum ampoule with a diameter of 8 mm and a height of 50 mm with a wall thickness of 0.2 mm. Then water was poured into the ampoule, the weight of which was determined by temperature and pressure of the run. The presence of water in the reaction ampoule after the experiment was considered a prerequisite for the reliability of the experiment. The welded ampoule was inserted into the Re-reactor, which was filled under pressure of 100 atm with argon-hydrogen mixture with a given molar fraction of hydrogen.

The samples were analyzed with a wide probe $(20-40 \,\mu\text{m})$ on the Tescan Vega II XMU scanning electron microscope (SEM) with energy dispersive (INCAx-sight) and wave (INCA wave 700) X-ray spectrometers. The location of the analyses in the figures is shown by symbol.

11.3 Research Results

11.3.1 Differentiation Types of Magmatic Complexes

The majority of differentiated magmatic complexes is located within the limits of ancient shields among deeply metamorphosed formations of the Archean or Proterozoic and have pre-Cambrian age (Bushveld, Stillwater, etc.). Less frequently stratified massifs date back to the Early Paleozoic age, being located on the activated margins of ancient platforms or in the zones of tectonomagmatic activation of consolidated median massifs. Single layered intrusions of later age up to tertiary (Skaergaard, Ram Island intrusions) are known. Regardless of age, at the early stages of development of folded regions, magmatism is represented by the effusives of komatiite or picrite-basalt composition, which in time are replaced by ophiolite plutonic complexes. Stratified differentiated stratiform massifs are formed in the consolidated areas of the Earth's crust, i.e. they are typical postorogenic formations associated with the stages of tectonic-magmatic activation of rigid structures. A special formation consists of differentiated trap intrusions formed under shallow depths in the effusive-sedimentary cover of the platforms.

Thus, magmatic complexes are characterized by different tectonic mode of formation and, as a consequence, were formed in different physicochemical conditions, as well as under the pressure of the magmatic fluid of different compositions, which directly determines the evolution of intrachamber melts, i.e. the type of differentiation. That is why stratified massifs belonging to the same type of differentiation, but formed in different geological epochs and in different regions (and even on different continents) have close and stable characteristics of structure and ore specialization (for example, massifs of Bushveld, South Africa, Stillwater, USA and the Fedorovo-Panskie tundras, Kola Peninsula). *Ophiolite complexes.* Geological formations of greenstone belts are the oldest rocks within the shields, having the features of ophiolite associations. Ore formations, which are spatially and genetically related to volcanosedimentary and intrusive complexes of komatiites, form in general a metallogenic specialization, typical for ophiolite complexes, of course, with their own peculiarities, probably due to the restorative regime of the processes of magmatism, metamorphism and ore formation, which took place at the early stages of the Earth's evolution. Components of this association are: bodies of ultrabasic composition containing chromite mineralization; intrusions of gabbroids with copper-nickel mineralization; formation and vein bodies of gold ore and copper-polymetallic (kies) deposits; in the main metavulcanoes of ferrous quartzite deposits (Watson 1980). In addition, the formational differences of the ancient greenstone belts are undoubtedly conditioned by the structures of the rift location (Grachev and Fedorovsky 1980; Moralev 1986).

In the full bottom-up ophiolite series the following types of rocks are presented: a complex of intrusive rocks consisting of harzburgites, lherzolites, wehrlites, dunites, peridotites, gabbroids and plagiogranites (Fig. 11.1); a complex of parallel dikes (sheeted complex) of the main composition, sometimes containing dikes and bodies of irregular shape of picrites and pyroxenites (perknites, Coish et al. 1982); volcanic complex, usually composed of pillow lava and more leucocratic ball-and-pillow rocks. The stratigraphic analogy of the ophiolite and oceanic crust has led many researchers to the concept of the oceanic origin of the ophiolite and the subsequent thrust to the continents in the subduction zones (Colman 1979). However, more detailed geological and petrological studies show their genetic differences. Still Miyashiro (1975) is tried to identify three types of ophiolite associations, relying on the petrochemical features of the volcanic series. Then such attempts were made



Fig. 11.1 Ophiolite complexes: (1a) Trends of differentiation of the Troodos massif (Cyprus). *I*—dunite-harzburgite basal zone; *II*—pyroxenite-anorthosite zone; *III*—gabbro-granitoids of the upper massif zone, sheeted dyke complex; *IV*—dunite-chromitite; Rocks of the massif: *I*—dunites, harzburgites, PI-peridotites; 2—PI-pyroxenites, anorthosites; *3*—ol-gabbros, Qtz-diorites, trondhjemites, tonalites, plagiogranites; *4*—magnetite ores; *5*—chromitite ores; (1b) Kimpersaisky massif. *I*—basal zone (dunite-garzburgite-lherzolite); *II*—lherzolite-anorthosite; *III*—supposed gabbro-plagiogranite; *IV*—separation of chromitite ores from dunite liquid. (1c) Jil-Satanakhachsky massif (Caucasus). *I*—basal zone (dunite-garzburgite-wehrlite); *II*—the middle zone (olivinite-troctolite-anorthosite); *III*—upper zone (ol-gabbro—gabbro—diorites—plagiogranites); *IV*—trend of the chromitite ores separation from dunite liquid



Fig. 11.2 Differentiation trends of oceanic rocks of the Atlantic Ocean. *I*—intrusive rocks of the mid-oceanic ridge and transform faults; *II*—picritic rocks; *III*—olivinite-plagioclase and plagioclase basalts (MORB)

repeatedly (Serri 1982; Beccaluva et al. 1983; Boudier and Nicolos 1985; Pierce et al. 1987). Now it has become clear that the set of rocks of mid-ocean ridges is one of the types of magmatic complexes, with its specific differences (Fig. 11.2). As the volcanic rocks of the mid-ocean ridges evolve, the picrite magmatism turns into a basaltic one, in which the anorthositic tendency of differentiation (MORB basalts, Fig. 11.2) manifests itself more typical for intrusive rocks. As the oceanic ridges consolidate, magmatism acquires a ferrous character, with the formation of Icelandic series that turn into liparites and complementary alkaline rocks. Intrusive rocks of mid-ocean ridges, as can be seen from the few samples associated with basalts, form a primitive olivine-pyroxene-plagioclase trend (I–II, Fig. 11.2).

A classic example of the plutonic phases of ophiolite complexes is the well-studied Troodos ophiolite complex in Cyprus, whose plutonic phase consists of three series of rocks that develop as if independently (Fig. 11.1a). Lower, dunite-harzburgite (*I*), up the section is replaced by pyroxenite-anorthozite rocks (*III*), which in turn are covered by gabbro, tonalites, trondjemites, plagiogranites (*IV*). All three series represent the ways of autonomous stratification of melts, probably related to each other by the primary trend of latent superliquidus nanocluster stratification. Petrochemically, diorites, diabases, gabbro-dolerites, and quartz-albite porphyries of the dyke complex coincide with both the upper gabbro-plagiogranitic series of the plutonic phase and with basalts, andesites, and dacites of the overlapping effusions. The dikes penetrate the upper part of the differentiated massif and intrude in volcanic rocks. From this position, intrusive formations can be considered as deep-seated chamber of volcanoes, and the study of the peculiarities of the formation and consolidation of massifs is directly related to the processes of magma evolution in volcanic series. If this is the case, it is the regularities of melts evolution in the upper part of magmatic chambers that determine the petrochemical characteristics of volcanic formations in combination with the intensity of tectonic movements that determine the periodicity of eruptions in time, as seen on petrochemical diagrams of plutonic complexes. As the magmatic chamber develops, eruption processes are likely to capture the melts of pyroxene—anorthosite trend, especially in the early stages of development of these rocks, forming dykes of appropriate composition, such as in the Betts Cove Ophiolite Complex, Canada (Coish et al. 1982).

As spreading evolves, dunite-garzburgite ophiolite complexes (Fig. 11.1a) occur from the oceanic side, dunite-garzburgite-lherzolite (Fig. 11.1b) and dunite-garzburgite-wehrlite (Fig. 11.1c) from the continental, and dunite-wehrliteclinopyroxenite massifs (Urals type of differentiation, Fig. 11.3) in folded belts transient to platform-type formations (Marakushev and Bezmen 1983). In more consolidated conditions, dyke belts degenerate and volcanic intensity decreases. In the plutonic phase, due to the more contrasting development of differentiation, the role of clinopyroxene-containing rocks increases, while the share of acid rocks (plagiogranites and tonalits) increases, the thickness of which is sometimes commensurate with the capacity of gabbroid rocks (for example, in the ophiolite complex Bail Mountain, in Appalachians, Coish et al. 1982). In this evolution, there is an increase in the accumulation of calcium, iron, titanium, and vanadium in magma, which is associated with the development of the alkaline trend of magmatism, which is peculiar to platforms.



Fig. 11.3 Urals type of differentiation. The Kytlymsky massif (Middle Urals). Trends of differentiation: *I*—dunite-clinopyroxenite; *II*—olivinite-troctolite-anorthosite; *III*—gabbro—granitoid; *IV*—*Ti-mag*-clinopyroxenites; *V*—*Ti-mag*-olivinites; *VI*—dunite-chromitite. Rocks of massif: *I*—dunites, wehrlites; 2—olivinites, troctolites, gabbro-troctolites, anorthosites; *3*—gabbro-norites; *4*—granitoids; *5*—*Ti*-magnetite clinopyroxenites; *6*—*Ti*-magnetite olivinites; *7*—*Ti*-magnetite ores; *8*—chromitite veins with nugget platinum

Urals type of differentiation. This type of differentiation (Fig. 11.3) shows a shift in rock compositions towards the development of anorthosites, troctolites, clinopyroxenites and associated magnetite ores. An example of dunite—clinopyroxenite formation is an ultrabasic belt in the Urals, in which intensive accumulation of iron in the troctolite (olivine-anorthozite) differentiation trend is traced petrochemically (*II*, Fig. 11.3) with the following decomposition of rocks into iron-rich and comparatively titanium-poor (up to 4% TiO₂) ores, Ti-magnetite bearing clinopyroxenites (*V*) and olivinites (*VI*, Fig. 11.3).

For experimental simulation of the gabbro-plagiogranite trend (*III*, Fig. 11.3), quartz gabbro glass was used. In order to avoid diffusion of iron into the platinum ampoule, the charge was pressed into a crucible made of Ni, Cr, Fe alloy. The experiments were maintained for 4 days at the pressure of $P_{total} = 4$ MPa, $X_{H2} = 0.5$, $X_{H2O} = 0.5$ and the temperature of 1150 °C, which is significantly higher than the liquidus temperature, which, as shown by our experiments, is below 950 °C at the water pressure equal to the partial pressure in the experiments with water-hydrogen fluids ($P_{total} = P_{H2O} = 200$ MPa).

The results of one of the experiments are shown in Fig. 11.4. During the experiment, three layers were formed (Fig. 11.4a): the upper, very narrow layer (1) of acidic composition (Qtz-diorite), the middle (2)—diorite and lower layer (3) of allivalite composition as fibrous-tangled clusters (Fig. 11.4b). The analysed compositions on the petrochemical diagram form a trend similar to the trend of differentiation of the upper series of Urals type massifs as well as the upper zones of ophiolite complexes and, as it will be shown below, of stratiform massifs.



Fig. 11.4 Experimental study of the troctolite-granitoid trend of evolution (Qtz-gabbro—starting composition). (4a)—photo of the sample after the experiment; (4b)—cluster aggregates (increase 1000^X); (4c)—petrochemical diagram of the results: 1—dacite layer; 2—matrix; 3—aggregates of clusters of allivalite composition



Fig. 11.5 Trends in differentiation of stratiform massifs. (5a) Bushveld Complex (South Africa). *I*—dunite—bronzitite; *II*—pyroxenite—anorthosite; *III*—granophyre—*Fe*-gabbro-magnetitite; *IV*—bronzitite—chromitite; Rocks of massif: *I*—dunites, harzburgites, bronzitities of the Basal zone; 2—PI-bronzitities, norites, anorthosites of the Critical and Main zones; 3—felsites, granites, diorites, ferrodiorites, gabbro, norites, melanogabbro of the Upper zone; 4—magnetite mineralization of the Upper zone; 5—chromitities of the Critical zone. (5b) Massifs: (white symbols) Stillwater (USA); (black symbols) Sudbury (Canada). *I*—dunite—bronzitite trend of the Stillwater ultrabasic series; *II*—bronzitite-anorthosite trend of the Stillwater Banded series; *III*—norite-granite trend of the Upper Sudbury zone; *IV*—chromitite ores in peridotite melts. (5c) Munni-Munni massif (Australia). *I*—dunite—wehrlite trend of the lower ultrabasic zone; *II*—pyroxenite-anorthozite trend of the Critical zone; *III*—Fe-gabbro-granitoid trend of the Upper zone

Stratiform massifs. From the point of view of completeness of the presented sections, the stratiform massifs located within the continental platforms and were developing in the absence of large tectonic movements. This led to the formation of intrusive complexes with a far gone differentiation from dunites to granites with the separation of ore magmas: sulfide, chromite, titanomagnetite. The most fully studied is the Bushveld complex, occupying about 29,000 km², timed to the most ancient deeply eroded structure of the South Africa shield type, formed in the Achean and then subjected to activation for a long time. The petrochemical diagram of this complex is shown in Fig. 11.5a.

The Basal zone (*I*) is composed of dunites, harzburgites, and bronzitites. The Critical series is characterized by the clearest stratification in comparison with other zones of the massif. In general, the Critical zone is characterized by a rhythmical alternation of bronzitites, norites, gabbronorites, anorthosites and chromites. The Main zone is the largest unit of the massif. In contrast to the previous zones, a thin rhythmic stratification was weakly manifested here. The rocks in general are represented by gabbro-norites and anorthosites. The petrochemical diagram of rocks, Critical and main zones form a pyroxene-anorthite trend (*II*). At the base of the Upper zone lies a horizon, which for the first time appears a significant amount of titanomagnetite, forming layers of monominal rocks, with a thickness of 0.3-1.8 m. Ferro-gabbro (ferro-diorites) prevail, which are replaced by thick layer of felsites and red Buschweld granites up the section. In general, the petrochemical diagram of the zone shows the granophyre—*Fe*-gabbro—*T*i-magnetite trend (*III*).

The calm tectonic situation contributed to the separation of chromite (IV, Fig. 11.5a), titanomagnetite (V, Fig. 11.5a), and sulfide composition ore magmas,

which are associated with PGE deposits (Naldrett 2003). The complex multi-trend structure of stratified basite-ultrabasite plutons with a far-gone differentiation is characterized by a complex formation. Chromite and copper-nickel sulfide mineralization are confined to the lower part (ultrabasic zones), sulfide PGE mineralization to the middle (pyroxene-anorthite) zone, and iron-titanium mineralization to the upper, ferrogabbro-anorthozite or ferrogabbro-granite zone, where rocks of the final stages of differentiation are concentrated.

The Stillwater and Sudbury stratiform massifs, which are well studied, are characterized by a similar type of differentiation (Fig. 11.5b). The first one in the modern erosion section is represented by its ultrabasic and main part, the second one—by rocks of the upper, granophyr-norite series.

Similar to the plutonic zones of the ophiolite complexes, which by their mineralogical and pentrochemical peculiarities form a series from dunite-garzburgite to dunite-garzburgite-wehrlite rocks, stratiform complexes are also characterized by formational diversity. In contrast to the Stillwater and Bushweld massifs, in which the ultrabasic rocks are represented by dunites, harzburgites, bronzitites and anorthosites, there are massifs with the composition corresponding to labradorites, in the structure of which the main role is played by lherzolites, wehrlite and more acidic anorthosites (andesinites). The representative of the latter is the Munni-Munni complex of Australia (Fig. 11.5c). This is a fairly large massif of about 225 km² with a capacity of 5500 m and lies among the Archean (3600–2650 million years) granite-green shale rocks of the Pilbara block of Eastern Australia and is 2925 million years old. Lowsulphide (2% wt% sulphides) disseminated PGE-like mineralization (up to 2.5 ppm) is confined to porphyrite plagioclase websterites and gabbro-norites and traced in the form of a reef with a thickness of several tens of meters over 12 km.

Compared to Bushveld massif, granophyrea rocks are almost completely absent in the Munni-Munni complex. Unfortunately, we do not have analyses of acid rocks confined to the tops of the section in the form of low-power layers, lenses, and schlieren accumulation in order to compare gabbro-granophyre differentiation trends (*III*, Fig. 11.5a).

The observed differences are probably determined not only by the initial gross composition of magma, but also by the peculiarities of differentiation. In the Bushveld massif in the process of evolution there was a contrasting separation of granophyre magmas with the formation of a powerful zone of upper granites, as well as the separation of anorthositic rocks enriched with calcium from magmas of the basite composition. In the Munni-Munni massif, differentiation was less contrasting, especially at the early stages of development (at the stage of latent stratification), which led eventually to the concentration of calcium in the form of clinopyroxene in the ultrabasic rocks, and sodium in anorthosites.

Skaergaard type. Among the stratified complexes, the Skaergaard massif in East Greenland is an excellent type of intra-chamber diffraction of magmatic melts. His petrochemical diagram is shown in the Fig. 11.6, which shows two trends: olivine-plagioclase (I) binding rocks of the main and ultrabasic composition and granophyr-ferrogabbro (II), reflecting the differentiation of detached magmas of acidic composition with complementary iron enriched melts.



Fig. 11.6 Scaergaard type of stratiform differentiation of massifs. Skaergaard (Greenland), Cap Edward Hill (Greenland), Rum Massif (island of Rum). Differentiation trends: *I*—dunite—anortosite stratified series; *I-UBZ*-Upper Border zone of the Skaergaard massif; *II*—Fe-pyroxenite-granophyre of the Upper zone

The Skaergaard massif is an example of layered plutons with a well exposed top of the section. In the majority of other intrusions of a similar type in the British-Arctic province (Belkheevsk, Ram, Sky, etc.), the lower parts of the sections are preserved, formed by the rhythmic interlayering of dunites and troctolites with gabbroids, which supplement the ultrabasic part of the olivine-plagioclase trend in the diagram (Fig. 11.6). Industrial concentrations of platinum and low-sulphide type gold have been found in the Scaergaard-type massifs (Nielson and Garmicott 1993). This type of differentiation is characteristic of the following massifs: Tsipring, Karelia, Talnakh, Siberia, Ioko-Dovyrensky, Transbaikalia, Volkovskoye, Ural, Kiglapait, Labrador.

Gabbro—anorthosite, gabbro—granite and gabbro—syenite massifs. These massifs are characterized by similar trends in the differentiation (Fig. 11.7). The trend of the Tzaginsky massif (1) form the following rocks: labradorites—troctolites gabbro-norites—gabbro—olivinites—pyroxenites—*Ti*-magnetite ores. The trend of the Chineisky massif (2) form the following rocks: anorthozites—gabbro—gabbronorites—*Ti-Mag*-pyroxenites—*Ti*-magnetite ores. The trend of the NovoMirgorodsky massif (3) form the following rocks: anorthozites—gabbro-anorthozites—gabbro—*Fe*-peridotites—*Ti*-magnetite ores. The trend of the Kalarsky massif (4) form the rocks: anorthozites—*Ilm* and *Ti-mag*-ores. The trend of the Northern Timan massif (5) form the rocks: nepheline syenites—syenites—gabbro—*Fe*-gabbro. The



Fig. 11.7 Layered gabbro—anorthosite, gabbro—syenite, gabbro—granite intrusion with ilmenite and *Ti*-magnetite ores. Massifs: Gabbro—anorthsite: (1–4): *1*—Tzaginsky, Kola Peninsula; 2—Chineisky, Transbaikal; 3—Novomirgorodsky, Ukraina; 4—Kalarsky, Siberia. Gabbro—syenite: 5—Northern Timan, Siberia. Gabbro—granite: Malyi Kuibas, Urals

trend of the Malyi Kuibas (6) form the rocks: granites—diorites—gabbro—gabbropyroxenites—*Ti-Mag-* ores. They were formed in the activation zones of the platforms in a relatively calm tectonic environment and are therefore contrasted with the separation of rich *Ti*-magnetite and ilmenite ores (up to 15% Ti).

As an example, we studied the superliquidus nanocluster differentiation of the melt of anorthosite gabbro (33 wt%) and ilmenite (67 wt%) composition of the Novomirgorodsky massif in Ukraine. Pyroxenites of this massif contain phenocrystals of atomic carbon, which testifies to the reducing environment of formation of the chosen massif rock association. The experiments were carried out at 1250 °C and at 400 MPa in a high gas pressure vessel. Homogeneous glass (210 mg) was pressed into a vitreous carbon crucible, which was placed into a 7 mm platinum ampoule. Then a solution (150 mg) of acids (HF—0.25 wt%, HCl—0.25 wt%, H₃PO₄—0.15 wt% and H₃BO₃—0.15 wt%) was poured into the ampoule and paraffin C_nH_{n+2} (50 mg) was added. The fluid phase composition was controlled by the molar fraction of hydrogen ($X_{H2} = 0.18$) in the argon—hydrogen cell of the reactor (Bezmen and Gorbachev 2017). Within 3 days the melt is layered into three compositions: ilmenite, rutile and anorthosite gabbro (Fig. 11.8).

Differentiated sills and traps. As well as for stratiform massifs, the formation of differentiated traps and sills occurred in a relatively calm tectonic environment, but the small size of intrusions does not allow to reach a high degree of differentiation, leading to the separation of anorthosite or granite melts.

The gabbro-dolerite traps are characterized by several types of differentiation. In layered subsurface gabbro-diorite-granophyre sills: Polysides, New Jersey, USA



Fig. 11.8 The result of experimental modeling of ilmenite ore formation in the Novomirgorodsky gabbro-anorthosite massif (Ukraine). (7a)—micro-photography of a stratified sample obtained as a result of melting of the primary homogeneous melt of the ilmenite-anorthosite composition (T = 1250 °C, P = 400 MPa). *I*—ilmenite melt; *II*—rutile melt layer; *III—Fe*-gabbro—anorthosite melt; (7b)—petrochemical diagram of experimental and natural data: 1—initial glass sample; 2-lower layer of *Fe*-gabbro—anorthosite composition (*III*); 3-upper zone (ilmenite, *I*); 4—rutile layer (*II*); 5—rocks of the Novomirgorodsky massif

(Pearce 1970; Shirley 1987); Anakinskiy sill, Lower Tunguska (Marakushev and Kaminsky 1990) and other olivine-containing gabbro-norites, gabbro, diorite and granophyres are united by olivine—plagioclase komatite-like differentiation trend, passing into enriched quartz rocks.

Layered Pechenga massif have a more complex two-zone structure. In the lower zone, olivinites, wehrlites, clinopyroxenites are developed, with which sulfide copper-nickel ores are associated. The upper part of the massif is represented by rocks of the basic and subalkaline composition. There are layers of Ti-magnetite clinopyroxenites and Ti-Mag-monomineral ores on the border of the two zones. Accordingly, the diagram (Fig. 11.9) shows two differentiation trends reflecting the complex intra-chamber evolution of the melts, first nano-clucters separation of the basic and ultrabasic zones, then their autonomous development with the concentration of sulfide ores in dunites and wehrlites melts of the lower zone and Ti-magnetite in plagioclase clinopyroxenite melt of the upper zone. It has been confirmed by experimental researches: influence of hydrogen-containing fluids ($P_{total} = 100$ MPa)



Fig. 11.9 Differentiation trends in stratified sills and traps. Comparative characteristics of differentiation trends of the Talnakh intrusion of the Norilsk district (Siberia) and Pilgujarvi massif (Kola Peninsula). (8a) Talnakh massif. I—peridotite—anorthosite trend; II—peridotite—diorite—granitoid (Skaergaard type of differentiation, see Fig. 11.6). (8b) Pilgujarvi massif. I—olivinite—peridotite trend of the Lower zone; II—pyroxenite—leucogabbro trend of the Middle zone of the massif; III—*Ti-Mag*-pyroxenite—essexite trend of the Upper zone; IV—*Ti-Mag*-pyroxenite trend of ferrous mineralization; V—Ni-Cu ores—peridotite trend of sulfide immiscibility. Rocks of the massif: 1—olivinites, peridotites, olivine pyroxenites; 2—plagioclase pyroxenites, gabbro, leukogabbro; 3—*Ti-Mag*-pyroxenites, gabbro, essexite; 4—*Ti*-magnetite ores, *Ti-Mag*-pyroxenites; 5—pentlandite—chalcopyrite ores in the olivinite and peridotite

on the melts corresponding on the bulk composition (Marakushev et al. 1984) and the upper zone of Pilgujarvi massif, Pechenga.

The traps of Pechenga and the Norilsk Region differ sharply in the type of differentiation (Fig. 11.9). Picrite dolerites, troctolites, olivine gabbrodolerites, gabbrodolerites of the most fully studied ore-bearing Talnakh massif form an olivineplagioclase trend complicated by the appearance of diorites and quartz diorites. In general, the trend of differentiation of the Norilsk massifs is similar to that of the Scaergaard type differentiation (Fig. 11.6), which is probably connected with the proximity of physicochemical conditions of formation, especially with respect to the oxidation-reduction regime.

11.4 Discussion of Results

The need to classify differentiated complexes is not in doubt, as the classification can serve as a basis for forecasting and searching for magmatic mineral deposits. Therefore, it is not by chance that the systematization of massifs was carried out repeatedly (Kuznetsov 1964; Naldrett and Cabri 1976; Besson et al. 1979; Belousov et al. 1982). However, either classification features external to magmatic bodies, such as connection with effusives, belonging to different tectonic structures, sizes

of intrusive bodies, etc., were chosen as the basis (Besson et al. 1979), or compositions of magmatic rocks in respect of individual oxides or groups of oxides. As a rule, the regularities of crystallization of fluid-free silicate petrologically important systems based on the physicochemical analysis of simple state diagrams based on experimental data in dry conditions were taken as the genetic basis of intrachamber differentiation. The petrochemical material presented in the article shows that, relying only on crystallization processes, it is impossible to deduce the whole set of rocks of magmatic complexes from granophyres to dunites, so it is no coincidence that hypotheses of multiple introduction and mixing of magmas in the chamber appeared (Irvine 1979; Campbell and Turner 1986; Naldrett et al. 1990, etc.). The multiphase magmatic introduction for most massifs is not supported by geological data. This process is contradicted by the following: in general, the natural change upward along the section of ultrabasic rocks of the main rocks, complicated by the rhythmic arrangement of the layers, parallel arrangement of the layers, if they crystallized before the manifestation of tectonic movements, the absence of numerous feeders, which we observe in the form of a layer of parallel dikes in the ophiolite complexes (sheeted dyke complex), with volcanic eruptions, as well as similarity in the structure of numerous massifs of one formation type. Besides, the difference in the composition of magma portions should be explained by some peculiarities of deep differentiation, which we do not see and have a weak hypothetical idea of, in addition, many of the phases of introduction should correspond to the compositions that are absent among effusive rocks.

At present, it is unlikely that anyone will deny the essential role of fluids in geological processes, including in magmatism. In terms of weight, the total number of fluid components dissolved in magma probably does not exceed 10%. Thus, for example, 5 wt% of water in basalt melt is 50–60 mol%, 0.5 wt% of hydrogen—60–70 mol%. Thus, magmatic melts in the majority, obviously, represent the fluid-silicate substance in which the silicate fraction makes a smaller part.

As our experimental studies (Bezmen 1992; Bezmen and Gorbachev 2014) have shown, nanoclusters can be separated in the Earth's gravitational field at superliquidus temperatures, forming initially cryptic stratification and then layers of different compositions in fluid magma. As a result of our experimental data it follows that the fluid melts of magmatic chambers are completely differentiated in the liquid state. Thus, the gravitational migration of different densities of nanoclusters in the magmatic chamber forms flotation, sedimentation and rhythmic types of melt stratification in the absence of temperature gradients in magma. Due to the migration of fluid and fluid-enriched clusters to the top of the massif, the crystallization processes are activated from the bottom to the roof of the magmatic chamber.

The developed diagram, which formed the basis for comparative petrochemical analysis of magmatic complexes, in an accessible and understandable form reflects the genetic features of the formation of massifs in the form of differentiation trends, the set and location of which in the diagram and were the main features of classification by type of differentiation.

It should be emphasized that massifs with a similar bulk composition are often characterized by different types of differentiation, which is difficult to explain from the standpoint of crystallization fractionation, as they are formed in close physicochemical conditions. As the rock varieties are determined by the dry crystallization with cotectics and eutectics, the melts similar in composition would have a single trend of differentiation, and the massifs would differ only in the quantitative ratios of rocks. It is necessary to emphasize that magmatic complexes were formed in the Earth's crust in a relatively narrow pressure range (100–500 MPa), so the fluids could not affect to the order of crystallization of minerals. In this connection, without involving the mechanism of pre-crystallization differentiation, it is impossible to explain the detected regularities of evolution of intrachamber differentiation of initial melts.

It should be noted that among the basic and ultrabasic rocks in the intrusive complexes in the regular interlaying there are also more acid rocks, up to silicites (Anhaeusser 1971), which are genetically related to the evolution of magmatic chambers. If this is the case, we are convinced once again that crystallization differentiation cannot be the only and universal mechanism of magmatic differentiation. It is the liquid separation of clusters that leads, as experimental studies show, to the liquid formation of acidic and ultrabasic melts in the differentiation of basite magma. Problems of relations between rocks of homodrome and antidrome sequences, as well as gradual transitions between them, often observed in sections, are easily explained from the standpoint of liquid differentiation. In general, more basic magmas can be formed in any part of magmatic chambers: everything depends on the ratio and content of fluid components in the fluid shells of nanoclusters (Bezmen 2001).

According to the location of trends on the diagram and the totality of the rocks composing them, the ophiolite intrusive complexes form subtypes, the family—from dunite-garzburgite to dunite-garzburgite-wehrlite. Common for all opolite massifs is the occurrence at the base of the section of interlaying dunites and harzburgites, sometimes with lenses or layers of chromites. It is believed that these are the restite, remaining from melting melts from the mantle and their subsequent transfer to the overlying horizons. Maybe it is. However, their layered structure, absence of high-bar minerals, and absence of direct signs of melting such as reverse zoning in minerals, and often gradual transitions with overlying rocks, testify in favor of their origin by magmatic differentiation.

Depending on the tectonic situation, as the Earth's crust consolidates, the role of clinopyroxene-containing rocks as well as lherzolites and wehrlites increases in the massifs. At the same time, the associations of overlying rocks are also naturally changing. In dunite-garzburgite complexes they are represented by pyroxeniteanorthozite rocks in dunite-garzburgite-wehrlite rocks—olivinites, troctolites and anorthosites. The corresponding trend of differentiation of the dunite-garzburgitelherzolite massifs occupies an intermediate position. In the upper parts of the section differentiation leads to the formation of tonalities and plagiogranites, the thickness of which is determined by the tectonic regime of massif formation. In a quieter environment, dyke belts degenerate, the thickness of effusive rocks decreases and the number of plagiogranite layers increases.

Ophiolite complexes are notable for the fact that they clearly show the genetic links between effusive and intrusive magmatism. The geological structure of ophiolite belts, the relationship between intrusive, subvolcanic and effusive rocks, and the comparison of their petrochemical characteristics (Fig. 11.1) show that the processes of differentiation in magmatic chambers in many cases determine the structure and composition of associated effusive differentiated complexes.

The dunite-pyroxenite massifs, as well as the ophiolite complexes, form belts shifted towards the continent in relation to the ophiolites (for example, the Urals platinum-bearing belt). They are characterized by a subplatform mode of formation, so they are often combined with alkaline rocks (syenites, granosyenites), which may be genetically related. Despite the absence of harzburgites, the duniteclinopyroxenite massifs correspond to the ophiolite-like type of differentiation in terms of the set of trends, but occupy a kind of extreme position in relation to them (Figs. 11.5 and 11.6). Compared to the ophiolite massifs, there is an intensive accumulation of iron in the troctolite (olivinite-anorthosite) differentiation trend with the subsequent formation of magmatic iron ores. The geochemical behavior of chromium is exactly the opposite. Large chromite deposits associated with dunite-garzburgite massifs (Troodos, Cyprus) and especially dunite-garzburgitelherzolite deposits (Kempirsaysky massif, South Ural) are replaced by poor chromitehercynite mineralization in dunite-garzburgite-wehrlite complexes, while in duniteclinopyroxenite massifs there is a vein chromite mineralization, confined to dunites. With regard to chromium, it has no industrial significance, but chromite-silicate separation of melts, as shown experimentally (Bezmen 1992), leads to the extraction of PGE into a chromite liquid, which crystallizes to form veins with native platinum. Later on, they served as a source of large placer deposits in the Urals, Siberia (Kondersky massif), Alaska and other regions of the world.

By their petrochemical peculiarities, stratiform complexes are divided into two types of differentiation: Bushveld (South Africa) and Skaergaard (Greenland). The first one combines three trends, the second one—two (Figs. 11.5 and 11.6).

Like ophiolite massifs, the Bushveldt type stratified complexes have a three-zone structure: the lower zone is Basal, composed of ultrabasic rocks of dunite (peridotite)pyroxenite series (dunites, peridotite, bronzitite, websterite), the middle, Critical zone is composed of pyroxenite-norite and anorthosite rocks and the Upper zone is composed of melanocratic gabbro and granophyres. They also form a family, which is reflected in the diagrams of shifting trends in differentiation. In ophiolite complexes, as the role of clinopyroxene increases, trends shift towards troctolite. Here the picture is reverse, as soon as bronzitises are replaced by lherzolites, wehrlites, websterites, rocks are saturated with silica, thus the composition of anorthosites changes from labradorites in the Bushveld massif, to andesinites in the complex Munni-Munni (Australia). It should be noted that in contrast to the ophiolite massifs, differentiation in stratiform complexes is more contrasting, which leads to the appearance of almost monominal pyroxenites and anorthosites. It is explained by the quieter tectonic situation and deeper conditions of formation. Long-term differentiation, first cluster formation, then crystallization, promotes the accumulation of fluids in certain parts of the section—as a result of which the following concentrations can be achieved when ore magmas are separated: chromite, sulfide with PGE, titanomagnetite (Bezmen 2001).

Stratiform complexes are remarkable in comparison with other types of stratified massifs, high concentrations of platinum and gold, genetically related to poor sulfide copper-nickel mineralization and chromitite, but their content is naturally decreasing with the increasing role of clinopyroxene.

Stratiform massifs of the Skaergaard type have a simpler structure. They are characterized by the troctolite (olivine-plagioclase) differentiation trend, which is complicated by the splitting off from granophyre magmas (Fig. 11.6).

The rocks composing trap formations also form several differentiation trends, but the small size of magmatic chambers and their rapid loss of volatiles do not lead to a contrasting separation of melts as in large stratified plutons.

Thus, the considered extensive petrological material testifies to numerous ways of melts evolution in layered magmatic complexes, the formation belonging of which is characterized by a set of differentiation trends associated with different parts of the section of magmatic chambers.

In connection with the consideration of the peculiarities of the evolution of magmatic complexes, the main question arises:—What is the reason for the diversity of types of differentiation? The line-up? Of course, this is an important factor, but the bulk compositions of many massifs, calculated even without taking into account the granophyres, which are often not taken into account, because they do not fit into the processes of crystallization differentiation, occupy a rather narrow field on the petrochemical diagram, corresponding to the main rocks. Indeed, if one does not take into account plagioclase basalts of mid-ocean ridges, which are widespread in young geological structures, picrite and tholeitic basalts, as well as basalts of island arcs, are limited to a narrow field coinciding with the compositions of differentiated magmatic complexes. It remains to be assumed that the fluid component mode is also the main factor of differentiation.

It has been shown experimentally (Bezmen 1992) that the composition of the fluid phase has a significant influence on the differentiation of the melts. Since petrogenic components have different chemical affinity to volatile elements, nanoclusters contain fluids of different compositions.

The presence of carbon-containing gases in the fluid phase, especially CH₄, stimulates the separation of ultramafic melts in the upper parts of the sample. As an example, we present the results of Ol–Pl stratification of the basal zone of the Bushveld complex in South Africa. The main role in this part of the section belongs to dunites and olivine bronzitites containing a small amount of plagioclase. The average composition of this zone is close to plagioclase-bearing olivine bronzitite. As the initial composition for the experimental simulation, the glass of the following composition was synthesized from oxides in the vacuum high-temperature furnace at 1700 °C: SiO₂-49.1; TiO₂-O.2; Al₂O₃-7.56; Fe₂O₃-9.48; FeO-5.36; MnO-0.16; MgO-22.79; CaO-4.52; K₂O-O.11; Na₂O-0.71. The experiments were carried out under the pressure of complex gas mixtures of H–O–C–S system at P = 400 MPa and T = 1350 °C. The molar fraction of hydrogen in the experiment was controlled by the argon-hydrogen mixture and amounted to $X_{H2} = 0.2$. The fugacities of other gases was controlled by the presence of elemental carbon from which the crucible was made and the melt of pyrrhotite. The fluid phase at experiment parameters had the

following composition: $X_{H2O} = 0.31$; $X_{H2} = 0.2$; $X_{H2S} = 0.003$; $X_{CO2} = 0.077$; $X_{CO} = 0.19$; $X_{CH4} = 0.218$; $\log f_{O2} = -9.9$; $\log f_{S2} = -3.71$. It is assumed that the given parameters are as close as possible to the conditions of formation of the Bushveldsky complex, in rocks of which elemental carbon is present (Touysinhthiphonexay et al. 1984), in gas-liquid inclusions of minerals reduced gases, including CH₄ (Ballhaus and Stushpfl 1985), in addition, oxidation-reduction conditions of formation of rocks are also close to natural (Elliott et al. 1982).

Under the pressure of hydrogen-containing fluids in the absence of a thermal gradient, the initial melt is stratified with accumulation of liquid close to the dunite in the upper zone of the crucible (Fig. 11.10). Super-liquid temperatures were confirmed by experiments under the pressure of pure water ($P_{total} = P_{H2O} = 120$ MPa), equal to the partial pressure in the mixtures ($X_{H2O} = 0.31$). After 3 days of experiment the upper peridotite zone is represented by large (up to 1 mm) crystallized olivine idiomorphic crystals, between which there is almost no intergranular liquid. This zone has the following composition: SiO₂-42.76; TiO₂-0.2; Al₂O₃-4.2; FeO-17.41; MnO-32.69; CaO-2.14; K₂O-0.007; Na₂O-0.44. The bottom zone, transparent glass,



Fig. 11.10 Super-liquidus nano-cluster differentiation of *Ol-Pl*-pyroxenite (example of antidromic stratification). (9a)—microphotography of the layered sample: upper zone, 1—peridotite, 2—*Pl*-bronzitite matrix, 3—bronzitite clusters aggregates, 4—pyrrhotite melt (sulphur fugacity indicator). (9b)—linear structure of nanocluster aggregates with large magnification (1000^x). (9c)—Petrochemical diagram of experimental data

has composition close to plagioclase bronzitite: SiO₂-50.80; TiO₂-O.2; Al₂O₃-9.99; FeO-14.34; MnO-0.15; MgO-17.61; CaO-5.78; K₂O-0.18; Na₂O-0.95.

In the lower zone there is a new liquid stratification (Fig. 11.10) in the form of packets or spheroids of accumulation of macromolecules, which are collected at the bottom of the sample, where they form a liquid enriched with bronzite: SiO_2 -51.09; TiO_2 -O.2; Al_2O_3 -9.37; FeO-15.13; MnO-0.15; MgO-18.36; CaO-4.71; K₂O-0.17; Na₂O-0.84. After quenching, it has a tangled fibrous structure, typical for hardened, colloidal, macromolecular liquids (Strepikheev and Derevitskaya 1976). In general, the initial composition of the fluid in the experiment determined the antidromic type of differentiation. Antidromic differentiation is characteristic of the Ural-Alaska, Elan-Vyazovsky and Podkolodnovsky massifs of the Voronezh crystalline shield and the Burpalinsky massif in Siberia. As a result of migration of volatile components to the upper part of the massif, the fluid composition changes, which affects the content and specialization of fluid components in the melts of cluster shells. As a result, in some parts of the section of stratified complexes, an antidromic type of differentiation appears (for example, in the Khibiny alkaline massif on the Kola Peninsula).

Unlike crystals or droplets of immiscible liquids that have phase interface boundaries, clusters or macromolecules are constantly exchanging individual molecules or parts of molecular groups with the matrix. These are fluctuating systems that do not have an interface and are permanently in dynamic equilibrium with the melt. This reduces the free energy of cluster formation, which prevents their unlimited increase in size, as opposed to crystal differentiation, which according to the laws of thermodynamics must constantly increase in size in magma by dissolving the smaller crystals and the growth of larger ones.

The interaction of clusters with the melt, the composition of which changes at the gravitational separation, can lead to a corresponding change at a constant temperature of the cluster composition (macromolecules), which we observe in the studied series of experiments.

The crystallization of ultrabasic melts raises separating fluids to higher horizons and interacts with overlying magma or partially or completely crystallized rocks, causing their melting and subsequent liquid differentiation (Fig. 11.11). Petrogenetically, this process is fixed by zones, often ore-bearing, with low temperatures of formation of finely stratified contrasting rocks, which are located in poorly differentiated strata.

The evolution of magmatism is associated with a restorative regime of magma formation, especially at the early stages of formation of magmatic chambers (Kadik et al. 1990; Bezmen 1992). At low chemical oxygen potential in the fluid phase, the share of hydrogen increases, which, interacting with magmatic melts, causes their intensive depolymerization (Bezmen 1992). In previous studies (Bezmen et al. 1991, 2005) it was shown that the presence of hydrogen leads to the breaking of the Si–O–Si bridge bonds in silica oxygen anions and the replacement of part of the oxygen atoms by hydrogen ions (fluorine, chlorine, phosphorus, boron) according to the following scheme:

$$-Si-O-Si + H = Si-O + Si-H.$$



Fig. 11.11 Photograph of outcrop in the upper stratified zone of the Panskiye tundra massif. Melt remelting and liquid differentiation of melts under the influence of ascending fluids. 1—Quaternary sediments, 2—spotted anorthosites, 3—gabbro-norites, 4—norites, 5—pyroxenites

According to the data of photoelectron spectroscopy in silicate melts even at low molar fractions of hydrogen ($X_{H2} \ge 0.1$) elementary silicon, which is probably formed during quenching, was found (Bezmen et al. 1991). Fluid-saturated magmatic melts produce fluctuating molecular groups-nanoclusters or nanomolecular groups, similar in structure to minerals, but containing hydrogen, water and other components, including ore ones. In the Earth's gravitational field, clusters move to form a cryptic and then contrasting stratification in the magmatic melts. Fluorine has a strong influence on the separation of melts enriched with anorthosite molecule, i.e. depending on its calcium potential it is concentrated in anorthosites or pyroxenites. Apparently, the composition of volatile components adequately influences the distribution of alkalis and silica in the cross-section of the massif, however, for the final conclusions about the influence of the fluid phase composition on the peculiarities of liquid differentiation of magmatic melts we need more targeted experimental data covering the natural range of fluid magmas, but in all cases it is necessary to have the presence of hydrogen in experiments or hydrogen-containing gases, CH₄, NH₃, HCl, H₂S, etc.

In general, massifs are characterized by simultaneous manifestation of the processes of liquid differentiation and crystallization in the recumbent of the massifs as the volatile components migrate to the upper part of the magmatic chamber. The wide development of evenly grained or fine-grained rocks, especially monominal rocks, in the sections of stratified massifs is almost unambiguous evidence of the predominance of the processes of superliquidus differentiation, while the origin of rocks with unevenly grained or taxitic structures may be due to the crystallization of magmas saturated with fluids.

11.5 Conclusions

The paper presents the petrochemical classification of differentiated magmatic complexes. For this purpose, a petrochemical diagram has been developed: Mg, Fe, Mn, Ca^{Px}, Cr, Ti–Na, K, Ca^{Pl}, Al–Si on which the main normative minerals are fixed by figurative points. As the main classification feature of magmatic complexes, the peculiarities of intrachamber evolution of magmatic melts were accepted.

The rocks of layered complexes are combined into differentiation trends, the totality and location of which determines the type of differentiation in the diagram. Within a single type, there is a shift in trends due to the composition of rocks of different massifs forming subtypes. Each differentiation trend has its own ore specialization. The type of differentiation determines the complex of ore mineralization of the massif.

The number and location of massif differentiation trends depends not only on the composition of the initial magmatic melts, but also on the physicochemical conditions of formation, with a special role to be played by the mode of fluid components.

Superliquidus differentiation of magmatic melts, crystallization of magmas at different levels of the section, separation of volatiles enriched with ore components, and their interaction with overlying magmas or rocks represent a complex petrogenetic system of evolution of magmatic chambers. At various stages of this process, fluid concentrations occur, leading to the separation of ore magmas, which are naturally confined to certain zones of the section of the massifs. Since the fluid components migrate both by themselves and in the fluid-containing magmatic shells of migrating clusters, stratification effects further develop under the influence of fluid inflow into the upper part of the chamber. In the lower parts of the magmatic chamber from bottom to top, the crystallization processes caused by the formation of solidus temperatures during the dissipation of fluid components from the melt are activated.

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