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Silicate inclusions in isoferroplatinum: Constraints on the origin of platinum mineralization in podiform chromitites

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ABSTRACT

The origin of large platinum-group mineral (PGM) nuggets inside chromitite schlieren in some Ural-Alaskan type ultramafic complexes has been assigned to orthomagmatic processes. A promising phenomenon which may provide new insights into the processes responsible for PGE mineralization formation, or at least place constraints on existing models, are the multiphase inclusions hosted in PGM which have been previously found in platinum nuggets from alluvial deposits. For the first time, this study examines multiphase inclusions hosted in isoferroplatinum nuggets from lode chromitite schlieren in almost unaltered dunite from the Matysken Ural-Alaskan type complex (Koryak Highlands, Far East Russia). These multiphase inclusions are comprised of diopside, hydrous silicates, apatite, plagioclase, K-feldspar, silica and other minerals that are distinctly different from the host dunite mineralogy. Taken together with similar mineral assemblages in Cr-spinel-hosted inclusions, these inclusions cannot be crystallization products of a mafic/ultramafic melt, but instead require an alternative explanation. We critically evaluate different genetic models and conclude that a range of fluid-assisted metamorphic processes should be involved in producing massive PGM within chromitite schlieren in dunites.

1. Introduction

Platinum-group elements (PGE) are among the rarest and most irregularly distributed elements in the Earth's mantle and crust, with concentrations barely reaching ppb levels in common rocks and climbing to ppm levels in economic deposits. It is widely believed that the most efficient mechanism of PGE transport and accumulation is with sulfide liquids for which these elements have a very strong affinity, where the efficient scavenging of PGE from mafic magmas into immiscible sulfide melts was noted in genetic models for orthomagmatic sulfide deposits, including Norilsk, Sudbury, etc. (Naldrett, 2004). The role of sulfide liquids in PGE accumulations was recently questioned by several discoveries of PGE nanoclusters, which formed before the melt became saturated in sulfur (González-Jiménez et al., 2019; Helmy et al., 2013), as well as the discovery of PGE alloys in high-temperature volcanic rocks (Kamenetsky et al., 2015). Furthermore, chromitites are sulfur-poor in the Ural-Alaskan mafic complexes (Johan, 2002; Tolstykh et al., 2004), which together with associated placer deposits, are a significant source of PGE. Aggregates of Pt-Fe alloys up to 150 g were reported in lode deposits within Ural-Alaskan type complexes in the Urals Mountains in Russia (Betekhtin, 1954), and nuggets up to 9 kg were found in the associated placers (see Orlov (2019)) and Fig. 1 for photos of historical museum samples).

The formation and high abundances of platinum-group minerals (PGM) in chromitites of Ural-Alaskan type complexes, especially in the absence of any evidence of PGE-bearing sulfide melts (Johan, 2002), remains enigmatic. Furthermore, the inhomogeneous distribution of PGM in largely dunitic rocks and their intimate association with disseminated lenses and schlieren of Cr-spinel further challenges the reigning orthomagmatic model.

A promising phenomenon which may provide new insights into the processes responsible for PGE mineralization, or at least place constraints on existing models, is the multiphase inclusions hosted in PGM. Such inclusions were described in PGM nuggets from alluvial deposits (Barkov et al., 2005; Cabri and Genkin, 1991; Dmitrenko and Mochalov, 1989; Mochalov, 2002; Johan, 2006; Nixon et al., 1990) and were shown to be typically composed of clinopyroxene and amphibole, along with subordinate albite, muscovite, quartz, and K-feldspar, and are thus are in stark contrast to any liquidus assemblage in common mafic magmas. However, previous studies of mineral inclusions in the alluvial PGM nuggets have established only hypothetical links for the origin of PGM nuggets and the possible source rocks in local peridotites,

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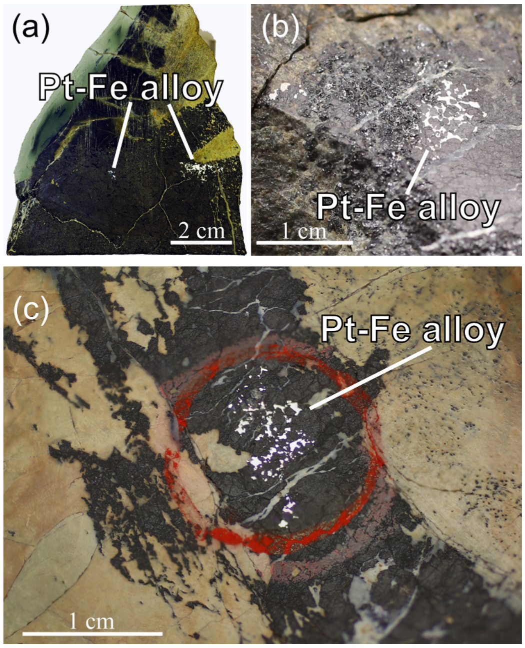

Fig. 1. Isoferroplatinum in dunite-hosted chromitite schlieren of Kamenushinsky (a) and Nizhnetagilsky (b, c) complexes (Urals, Russia). Samples are from the collections of Sergey Stepanov (a) and VSEGEI Geological Museum, Saint-Petersburg, Russia (b, c).

thereby rendering genetic constraints tenuous. The only available data on such inclusions in lode PGM is a report of single clinopyroxene-mica inclusion in the isoferroplatinum of the Galmoenan complex (Koryak Highlands, Russia) (Nazimova et al., 2011).

In this paper, we present the first detailed study of numerous multiphase inclusions in Pt-Fe alloys intergrown with Cr-spinel found in lode samples in dunite from a Ural-Alaskan-type ultramafic complex and compare this data with existing models of PGE mineralization.

2. Geological background

2.1. General information on Ural-Alaskan type complexes

It is well established that Ural-Alaskan type zonal complexes are the source of PGM for platinum placers, such as the placer deposits of the Urals Platinum Belt (Malitch and Badanina, 2015; Palamarchuk et al., 2017; Razin, 1976; Stepanov et al., 2017, 2019; Tolstykh et al., 2011; Zaccarini et al., 2018), the Koryak-Kamchatka Platinum Belt, including the Galmoenan placers (Russia) (Batanova et al., 1991, 2005, Batanova and Astrakhantsev, 1992, 1994; Kutyrev et al., 2018; Nazimova et al., 2011; Sidorov et al., 2019; N. D. Tolstykh et al., 2002; N. Tolstykh et al., 2002; Tolstykh et al., 2000, 2004), Goodnews Bay in Alaska (Foley et al., 1997; Tolstykh et al., 2002), Tulameen in British Columbia (Barkov et al., 2005; Nixon et al., 1990), and Colombia (Tistl et al., 1994). Several previous works showed that the main source for these placers are chromitites, which are located in the central dunitic zone of the massifs (Kutyev et al. 1991; Johan 2002; Tolstykh et al. 2004; Augé et al. 2005; O'Driscoll and Gonzales-Jimenez, 2014).

In the addition to being the source of economic-grade PGM placers, Ural-Alaskan type complexes have the following key features:

(1) Zonal structure, which is composed of a dunite core that grades through to wehrlites and clinopyroxenites, and then into gabbro (Batanova et al., 2005, 1992; Johan, 2002; O'Driscoll and González-Jiménez, 2016);
(2) Absence of orthopyroxene-bearing rocks such as harzburgites, lherzolites and norites, which is the main difference between Ural-Alaskan type and ophiolitic complexes (e.g. O'Driscoll and González-Jiménez, 2016);
(3) Presence of podiform chromitites (e.g. Augé et al., 2005; O'Driscoll and González-Jiménez, 2016);
(4) The predominance of Pt-Fe alloys over other PGM in both placer and lode occurrences (Augé et al., 2005; Tolstykh et al., 2015; O'Driscoll and González-Jiménez, 2016; Stepanov et al., 2019).

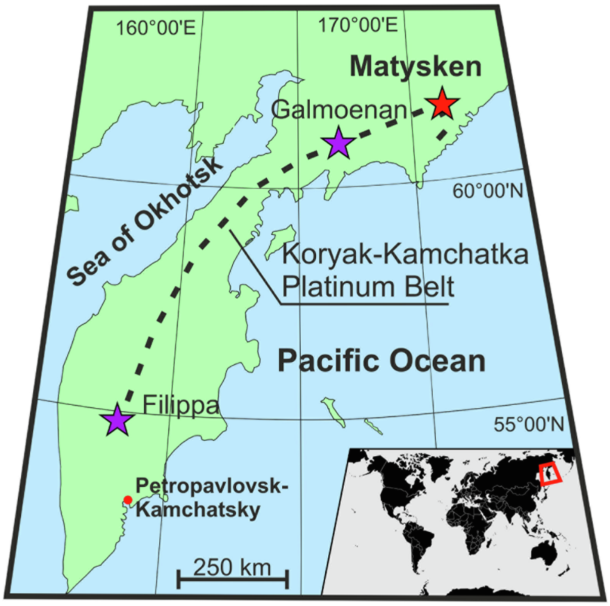

Fig. 2. Position of Koryak-Kamchatka Platinum Belt and Matysken complex in Kamchatka region and world (inset).

2.2. Koryak-Kamchatka platiniferous Belt

The Koryak-Kamchatka platiniferous belt extends approximately 1300 km longitudinally from the Filippa complex in the south of the Kamchatka Peninsula to the Matysken (another possible name is "Snegovoy") complex in the center of the Koryak Highlands (Fig. 2). The Ural-Alaskan type complexes are located in the Upper-Cretaceous units of Olyutor terrane (Batanova and Astrakhantsev, 1994). The basal unit of this terrane is the Vatyn formation which is composed of pillow basalts with MORB geochemical signatures that are interbedded with radiolarian-bearing cherts red jaspers and hyaloclastites. The upper unit is the Achayvayam formation which is composed of basaltic and picritic volcanics with arc geochemical signatures and is associated with pyroclastic, tuffites, sandstones and siltstones, red jaspers and hyaloclastites. The lower suite is interpreted to be oceanic crust, while the upper is a product of subsequent arc volcanism (Batanova and Astrakhantsev, 1992, 1994).

2.3. Geology of the Matysken complex

The Matysken complex contains a dunite core (area of $\approx 1.5–2.0$ km^2) with a thin outer zone of wehrlite and clinopyroxenite (Fig. 3b). The boundaries between ultramafic units are sharp and usually represented by zones of serpentinites and serpentinite breccias (Batanova and Astrakhantsev, 1992). The thickness of such zones varies from 10 cm to several tens of meters. All contacts between ultramafic units and host rocks of the Vatynskaya formation are tectonic (i.e. no quenched margins were observed). The only unit with direct contacts with the host rocks is gabbro, which is distributed at the outermost parts of the complex and sometimes forms thin dykes in the dunite (Fig. 3).

Dunites in the Matysken complex are not layered, fairly unaltered

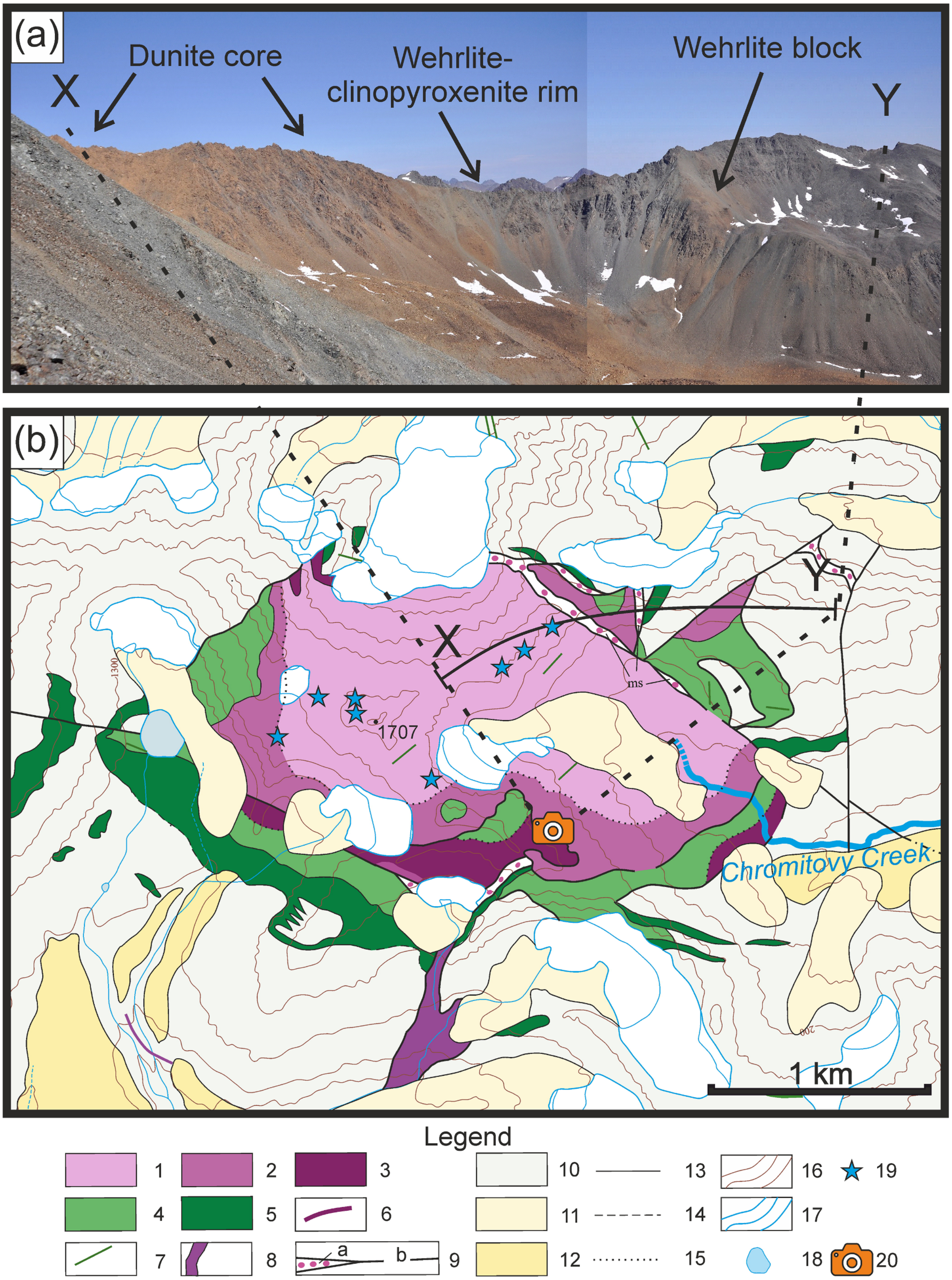

Fig. 3. Photo of Matysken massif outcrop (a) related to its geological scheme (b) 1 –dunite, 2 – dunite + wehrlite, 3 – wehrlite, 4 – olivine clinopyroxenite and clinopyroxenite, 5 – gabbro, 6 – picrite dykes, 7 – gabbro dykes, 8 – olivinite, 9 – serpentinization zones, a – in scale, b – out of scale, 10 – volcanic and sedimentary rocks of Late Cretacous Vatynskaya formation, 11 – glacial sediments, 12 – alluvium, 13–15 – geological borders (13 – visible sharp contacts, 14 – estimated contacts, 15 – facial transition), 16 – contour relief, 17 – glaciers, 18 – lakes, 19 – selected sampling points, 20 – point from which the photo “a” has been taken. Modified after unpublished data of A.V. Razumny.

and mainly consist of high-Mg olivine ($Fo_{0.88\text{-}0.92}$; Fo = Mg/(Mg + Fe^{2+}) × 100) with accessory diopside and chromite (Fig. 4c, d) (Batanova and Astrakhantsev, 1992). Zones containing highly altered lithologies are also present, however, there is no apparent relationship between alteration grade and the abundance of chromitites or platinum mineralization. For example, sample “M420” (Fig. 4c) is the most enriched in PGM and contains 0.5 wt% of OH^-.

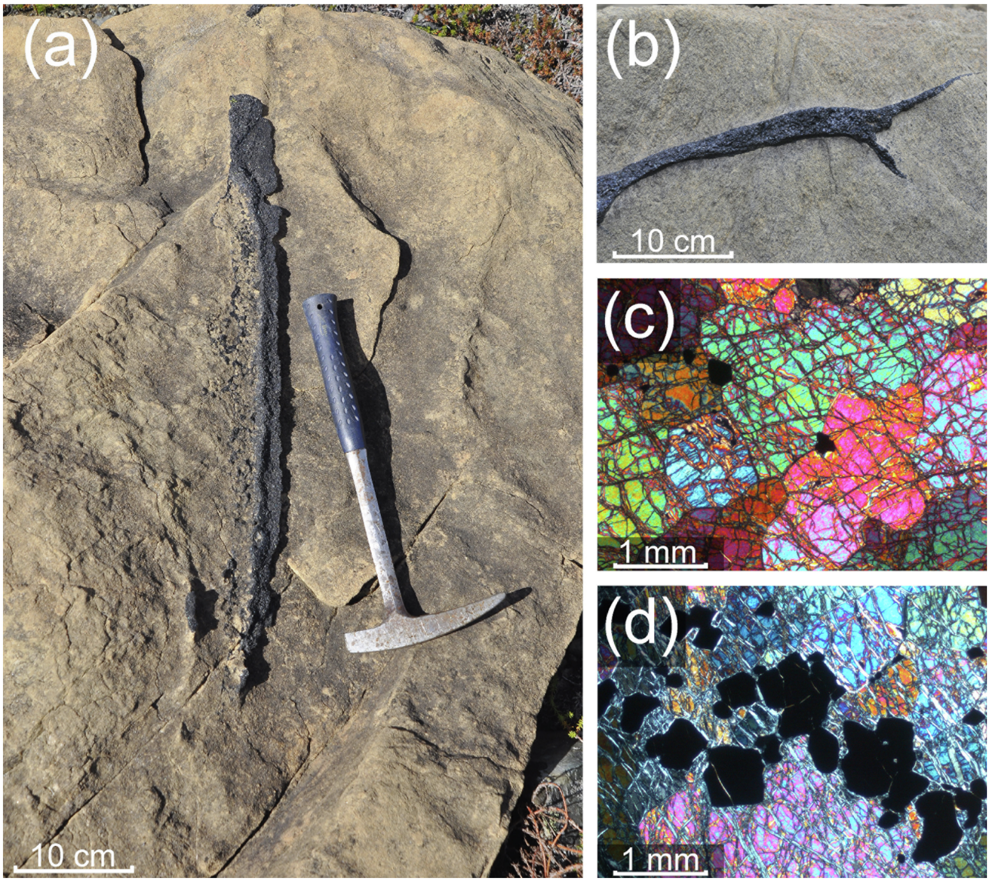

Fig. 4. (a, b) – outcrops of typical PGM-bearing chromitites of the Matysken complex, (c) – thin section of sample M420, one of the samples where large aggregates of PGM were found, (d) – thin section of the outer part of chromitite vein where it wedges out and gradually transitions into the Cr-spinel-rich dunite. (d, c) – polarized light.

3. Methods

3.1. Sampling and preparation technique

This study is based on the results of field work, which was conducted in 2016–2017. More than 300 samples of dunite and chromitites were collected. Ordinary samples weigh between 1.0 and 1.5 kg, and in the case of rocks containing visible chromite mineralization, samples were between 5 and 10 kg. After collection, samples were divided into two parts. The first part was sliced into multiple 1–2 mm sections with a diamond saw. This method assisted in finding large isoferroplatinum aggregates (> 1.0 mm), which were visible even before polishing. After polishing, numerous PGM between 1 and 250 µm in size were visible. The second part of the samples were crushed into particles between 0.5 and 1.0 mm in size and then panned in order to separate heavy mineral concentrates, which were then studied by stereomicroscope and PGM grains were manually extracted.

3.2. Analytical techniques

Polished samples, as well as PGM grains, were investigated in the Institute of Volcanology and Seismology FEB RAS by backscattered electron (BSE) imaging. Compositions of small silicate inclusions were studied by energy dispersive X-Ray spectroscopy using a Tescan VEGA-3 system equipped with an Oxford XMax80 EDS detector. A beam accelerating voltage of 20 kV, 20 s counting live time and 0.7nA current intensity was used. The following analytical lines of X-ray spectra were used to detect elements: Mα lines—for Pt, Os, and Ir; Lα lines—for Ru, Rh, Pd, and As; and Kα lines—for Fe, Cu, S, Ni, Al, Mg, Ca, V, Mn, Ti, Cr, and O. The following standards were used: pure metallic Pt, Os, Ir, Ru, Rh and Pd for PGE, sanidine for Si, K and Na, blue diopside for Ca, MgO for Mg, Al_2O_3 for Al, TiO_2 for Ti, $AlPO_4$ for P, V_2O_5 for V, rhodonite for Mn, FeS_2 for Fe and S, pure metallic for Ni. Electron backscattered diffraction (EBSD) on a Hitachi SU-70 SEM was used to evaluate the crystallographic orientation of the crystal microstructures. Ion-polishing for EBSD studies was done in Adelaide Microscopy. The EBSD analyses were performed using an acceleration voltage of 20 kV and ~3nA beam current using an Oxford AZtec NordlysNano EBSD detector integrated with an EDS system at University of Tasmania.

4. Results

4.1. Platinum mineralization of the Matysken complex

The vast majority of PGM occur in chromitite schlieren in dunite (Fig. 4a, b). The width of Cr-spinel accumulations does not exceed 10 cm, while their length is restricted to 1–2 m. This schlieren wedge out sharply (Fig. 4a), bifurcate (Fig. 4b) and/or disperse into disseminated clusters of Cr-spinel grains (Fig. 4d).

PGM were found in heavy mineral concentrates of crushed chromitites (Fig. 5a, b, e) and *in-situ* in polished sections (Fig. 5c, d). Small PGM grains a few micrometers in size can be found in most chromitite samples, whereas aggregates > 30 µm are uncommon, and “visible” PGM larger than 250 µm are rare. In general, platinum mineralization of the Matysken complex is similar to those of other Ural-Alaskan complexes worldwide (e.g. Tolstykh et al., 2004; Stepanov et al., 2019). The main PGE phase is isoferroplatinum, which is typically intergrown with Fe-rich Cr-spinel. Textural observations and EBSD analyses show that large aggregates of isoferroplatinum are composed of multiple subhedral crystals cementing euhedral Cr-spinel grains (Fig. 6). Direct contacts of isoferroplatinum with olivine are extremely rare. A unique case is shown in Fig. 4e, where intergrown olivine and euhedral Cr-spinel are included in isoferroplatinum. The most abundant phases after isoferroplatinum are Os-Ir-Ru alloys (Fig. 5f-h) and erlichmanite-laurite

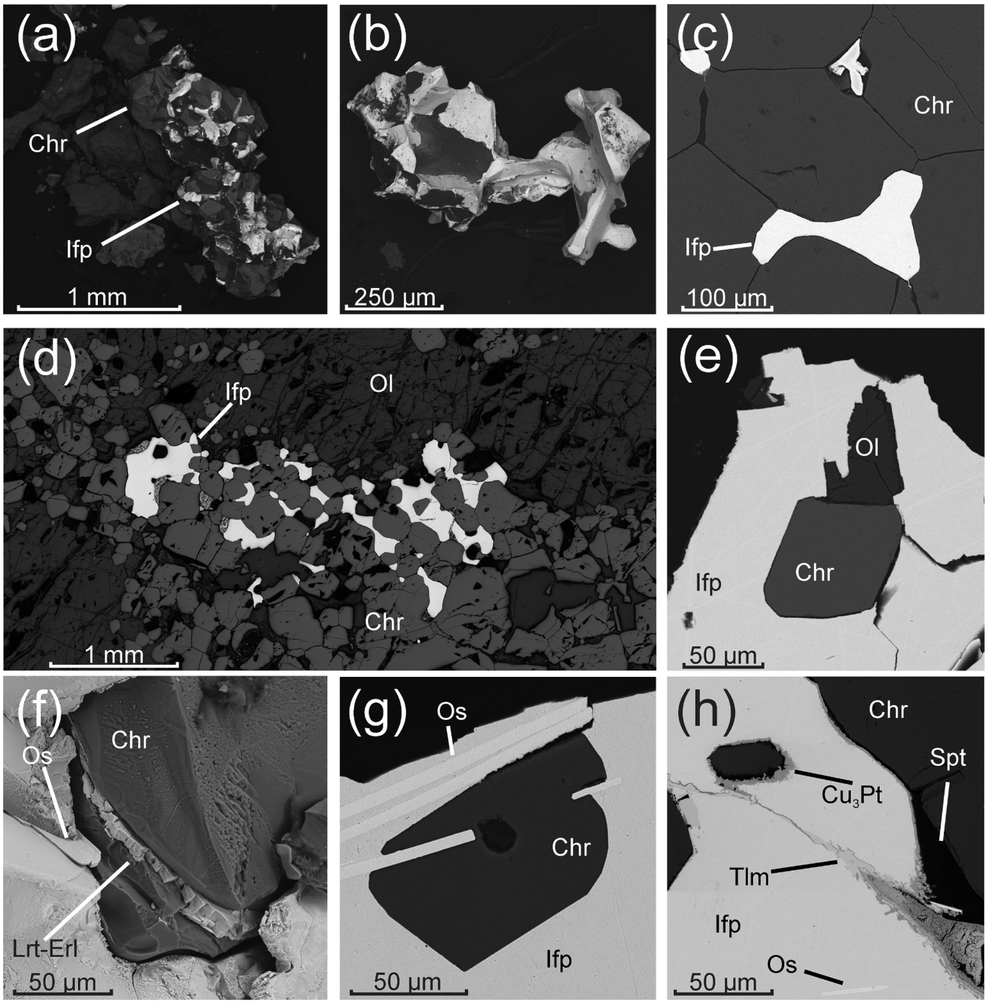

Fig. 5. Cr-spinel-isoferroplatinum aggregates from Matysken complex dunite: (a) aggregate of isoferroplatinum (Ifp) and Cr-spinel (Chr) from a heavy mineral concentrate, (b) – triple junction at the contact between Cr-spinel grains in the chromitite. Note the change in Cr-spinel shape at the contact with isoferroplatinum, (c) euhedral inclusions of native osmium (Os) at the contact of Cr-spinel and isoferroplatinum, (d) – isoferroplatinum-Cr-spinel aggregate in the polished dunite sample, (e) – inclusion containing Cr-spinel and olivine (Ol) in isoferroplatinum, (f) – isoferroplatinum and Cr-spinel intergrowth, the lamellae of native osmium and a planar inclusion of laurite-erlichmanite series linear (Lrt-Erl) may be noted, (g) – inclusion of native osmium and Cr-spinel in isoferroplatinum, note the silicate inclusion at the central part of Cr-spinel grain, (h) – intergrowth of isoferroplatinum with Cr-spinel and serpentine (Spt), note the secondary PGM such as tulameenite (Tlm) and unnamed mineral Cu_3Pt along fractures in isoferroplatinum. Image (d) – reflected light, other – BSE.

solid solutions (Fig. 5f). Secondary minerals that usually replace isoferroplatinum in the Ural-Alaskan type complexes, such as tetraferroplatinum, tulameenite or arsenides (Augé et al., 2005; Cabri and Genkin, 1991; O'Driscoll and González-Jiménez, 2016; Tolstykh et al., 2002, 2004, 2015), are also present in some samples (Fig. 5f), but are very rare in the isoferroplatinum grains which host inclusions, and absent from the inclusions themselves. The compositions of the most abundant PGM are listed at Supplementary Table S1.

4.2. Multiphase inclusions in isoferroplatinum

Forty-five multiphase inclusions in isoferroplatinum were studied. The distribution of inclusions in isoferroplatinum is uneven, where inclusions are absent from isoferroplatinum grains on some polished surfaces whereas in other grains multiple inclusions were observed (Fig. 7a). Inclusions in isoferroplatinum are between 5 and 50 μm in size (Fig. 7), their shape is usually polygonal or close to rectangular, and if several inclusions occur in the same nugget, they exhibit parallel aligned outlines.

Isoferroplatinum-hosted inclusions are typically composed of diopside and pargasite (Fig. 7). At least one of these phases occurs in every inclusion and is usually the dominant mineral in size (Fig. 7b–f). The inclusions also contain Ab-rich plagioclase ($An_{0.01}$-$_{0.15}$), K-feldspar, muscovite, phlogopite, apatite and titanite. In one of the inclusions, a small SiO_2 grain was observed. The shape of most hosted minerals appears to be controlled by the outlines of the cavity (Fig. 7). Diopside and pargasite within the inclusions are commonly euhedral, compared to muscovite and phlogopite, while plagioclase and K-feldspar are interstitial to all other minerals.

Crystals of laurite (RuS_2) and kashinite (Ir_2S_3) are present in approximately 50% of inclusions. Kashinite is subhedral to the host isoferroplatinum (Figs. 7b, d, e, 8) and thus seems to be a part of the inclusion assemblage. Laurite tends to form euhedral-shaped inclusions in isoferroplatinum that "penetrate" the inclusion outlines (Fig. 7b, e). Native osmium forms thin lamellae that are euhedral relative to all other minerals and sometimes determine the habit of the entire inclusion (Fig. 7c, d, f). Cr-spinel typically occurs as individual grains in isoferroplatinum and was documented only once in a multiphase inclusion (Fig. 7e).

The parallel alignment of the inclusion boundaries leads to the assumption that the inclusions could represent negative crystals whose shape is determined by the crystallographic orientation of host isoferroplatinum. This was tested by EBSD (e.g., Fig. 9a, which shows an isoferroplatinum grain containing numerous inclusions (see also Fig. 8)) and revealed that this grain consists of two crystals: i) the major one, which hosts the inclusions, and ii) the smaller part, which has a different orientation (Fig. 9b). The fact that all of the inclusions in Fig. 8 occur in monocrystalline isoferroplatinum supports the idea of them being negative crystal shaped. However, the complex orientation of the crystal with respect to the plane of the samples polished surface does not allow us to resolve this problem univocally.

4.3. Multiphase inclusions in Cr-spinel

Multiphase inclusions in Cr-spinel are common in all studied samples, including those with massive isoferroplatinum. Their size ranges widely from 5 to 80 μm (Fig. 10). Similar to inclusions in

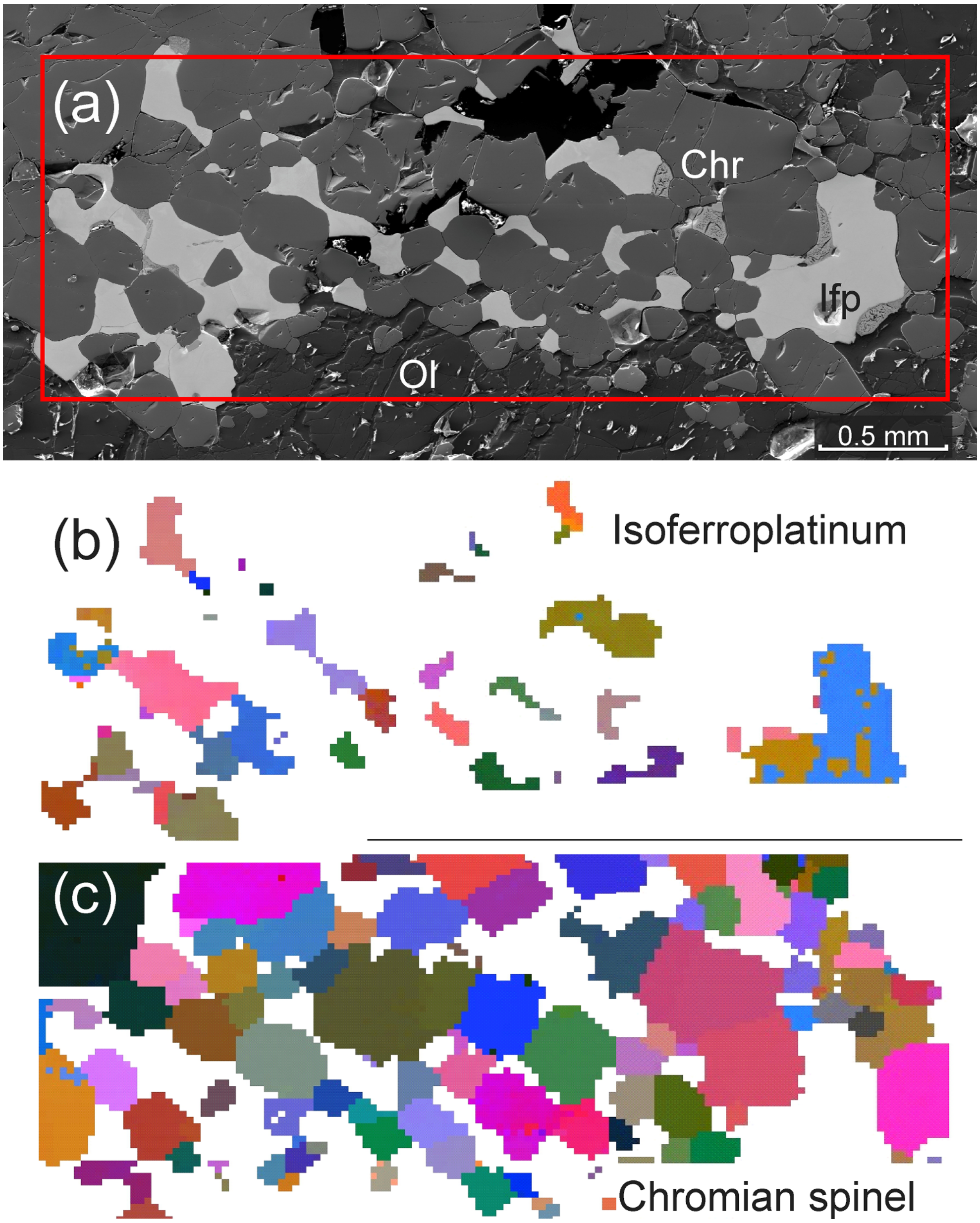

Fig. 6. EBSD analysis of Cr-spinel-isoferroplatinum aggregate from Fig. 5d: (a) – BSE + SE, (b) – isoferroplatinum in Euler colours, (c) Cr-spinel in Euler colours. See Supplementary Table S2 for more EBSD data.

isoferroplatinum, Cr-spinel-hosted inclusions are commonly composed of diopside, pargasite, apatite, and phlogopite, whereas plagioclase, feldspar, and silica are absent (Figs. 7 and 8). Furthermore, Cr-spinel contains garnet (andradite and/or uvarovite), chlorite, perovskite, serpentine and base metal sulfides that also contain multiphase inclusions (Figs. 7 and 8). The textural relationships between minerals in the Cr-spinel-hosted inclusions are also similar to those in the isoferroplatinum-hosted inclusions.

5. Discussion

5.1. The relationship of multiphase inclusions and platinum mineralization

Previously, isoferroplatinum-hosted multiphase inclusions were found in several localities (Table 1):

(1) Placers related to the Tulameen Ural-Alaskan-type complex (Nixon et al., 1990);
(2) Placer deposits in British Columbia (Barkov et al., 2005);
(3) Placers related to ophiolites of the Koryak Highlands (Dmitrenko and Mochalov, 1989; Mochalov, 2002);
(4) Placer deposits related to the Aldan-type Kondyor complex (Shcheka et al., 2004);
(5) Placer deposits of the Ural Mountains (Cabri and Genkin, 1991; Johan, 2006);
(6) Placer occurrences related to the Itchaivayamsky complex in the Koryak Highlands (Sidorov et al., 2019).
(7) The only occurrence of inclusions in lode isoferroplatinum is described as a single mica-clinopyroxene inclusion from the chromitite of Galmoenan Ural-Alaskan type complex in the Koryak Highlands (Nazimova et al., 2011).

The lack of data on multiphase inclusions in lode samples has led to uncertainties in understanding genetic relations between PGM nuggets and their source rocks, and thus complicated interpretations of primary nature of hosted multiphase inclusions. For example, Mochalov (2002)

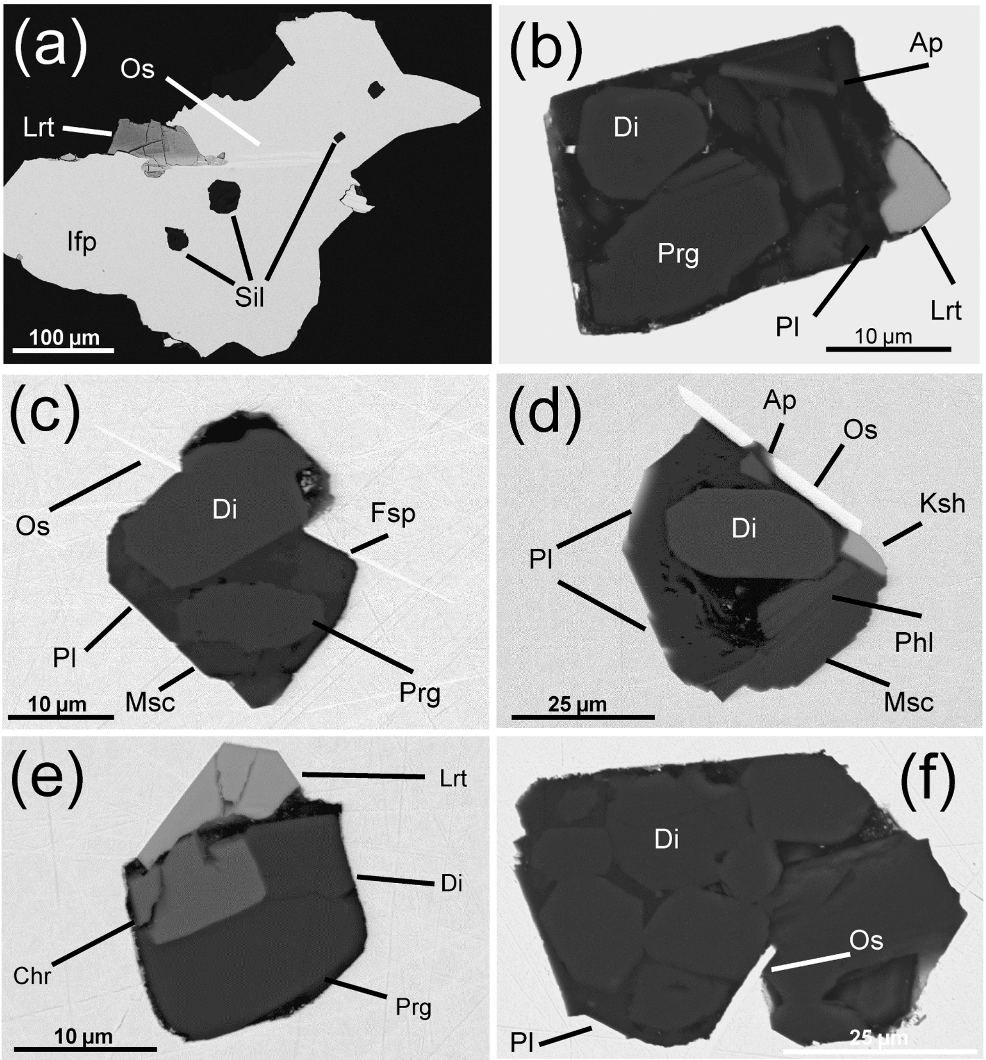

Fig. 7. Multiphase inclusions in isoferroplatinum. Mineral abbreviations: Os – native osmium, Di – diopside, Sil – multiphase inclusions, Prg – pargasite, Ap – apatite, Lrt – laurite, Fsp – K-feldspar, Pl – plagioclase, Ksh – kashinite, Msc – muscovite, Phl – phlogopite, Chr – Cr-spinel. See Supplementary Fig. S1 for more isoferroplatinum-hosted inclusions.

proposed that quartz, albite and K-feldspar hosted by inclusions in isoferroplatinum are of “clastogenic” origin (i.e. they are a result of mechanical entrapment of detritus minerals during the transportation of nuggets in rivers), while other minerals were interpreted to be the result of a post-magmatic fluid-assisted process. Other authors interpreted inclusions of quartz in native osmium as a sign that it originated from an olivine-free rock: “*The presence of euhedral quartz enclosed within the Ir–Os alloy, which probably crystallized from a trapped intercumulus liquid, is consistent with a mafic source… and is not consistent with dunite or another rock containing olivine*” (Barkov et al., 2005, page 1705). However, our study demonstrates that in case of Matysken complex, these minerals, being rare or absent in common ultramafic rocks, but present in the studied isoferroplatinum, must be closely related to the alloy's formation in dunite. This is supported by (a) *in-situ* occurrence of PGM among Cr-spinel grains, thereby negating the idea that felsic minerals may indicate a non-dunitic source of PGM; (b) a low alteration level of dunite and absence of any signs of interaction by external fluids (e.g. granitic intrusion), (c) an absence of any signs of isoferroplatinum alteration along the contacts with the inclusions (tetraferroplatinum, tulameenite or arsenides) that have been routinely found in some Ural-Alaskan complexes (Augé et al., 2005; Cabri and Genkin, 1991; Kutyrev et al., 2018; Malitch et al., 2017; O'Driscoll and González-Jiménez, 2016; Tolstykh et al., 2002, 2004, 2015), and (d) the absence of fractures transecting inclusions, which could be interpreted as potential pathways for infiltrating fluids (Fig. 7).

Thus, multiphase inclusions are directly related to the process of PGE accumulation and PGM formation and should be used as a cornerstone for genetic constraints.

5.2. Comparison of multiphase inclusions in isoferroplatinum and Cr-spinel

Studies of isoferroplatinum-hosted multiphase inclusions are scarce. On the other hand, the models of podiform chromitite formation based on Cr-spinel-hosted inclusions are much more abundant (Borisova et al., 2012; Pushkarev et al., 2007; Rollinson et al., 2018; Tolstykh et al., 2019, Zagrtdenov et al., 2018), largely due to the relative simplicity in obtaining samples. Thus, the application of the proposed model for Cr-spinel-hosted inclusions for explaining those in isoferroplatinum seems to be one of the simplest ways for understanding their nature. Comparison between inclusions in platinum and Cr-spinel coexisting in the same rock and same chromitite schlieren showed that they have similar morphologic features (compare Figs. 7 and 10). Another similarity is that the minerals which are either rare or totally absent in host dunites are the main constituents of these inclusions (Fig. 11). However, a more detailed comparison of the inclusions mineralogy revealed principal differences in their composition:

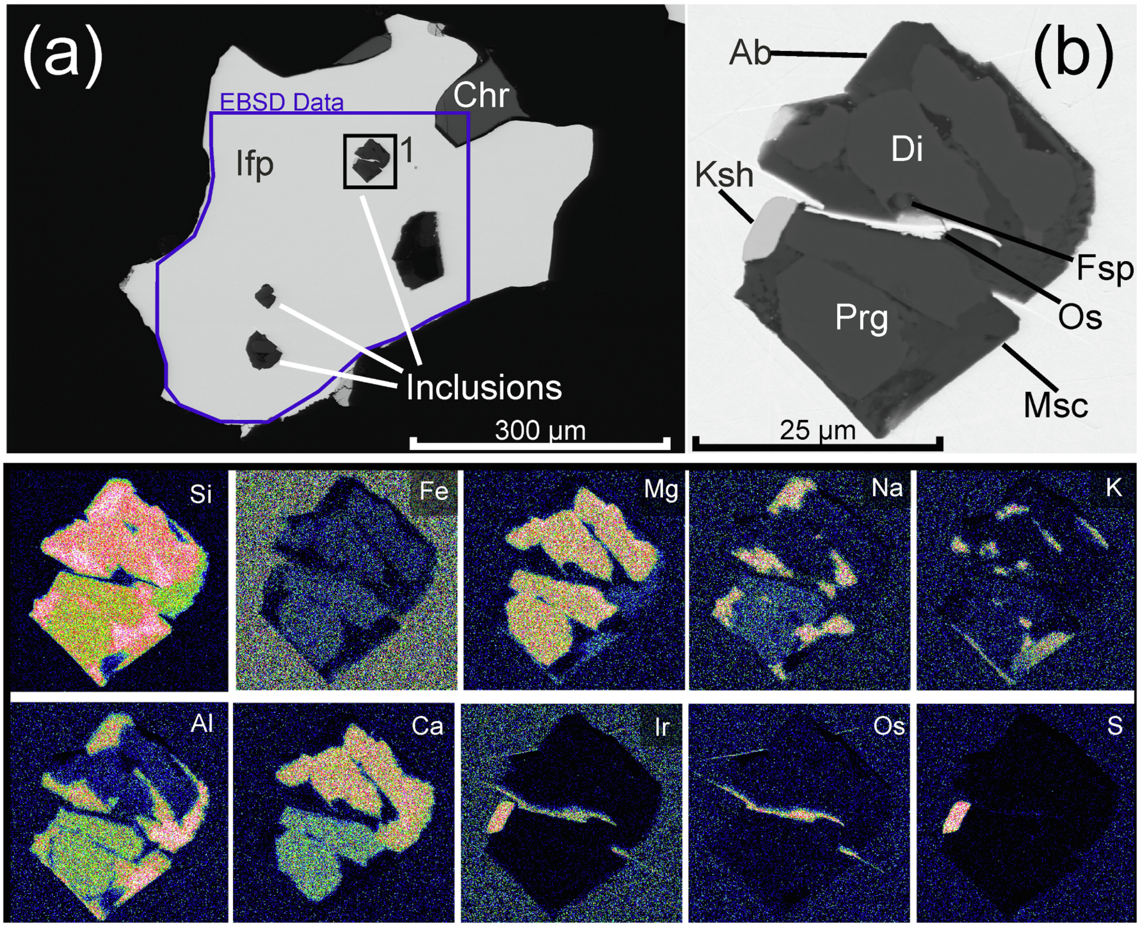

Fig. 8. X-ray element distribution maps of the multiphase inclusion in the isoferroplatinum: (a) general view of a grain, with marked area of EBSD study (see Fig. 9), rectangle “1″ marks the inclusion from figure (b) and maps. More element distribution maps for multiphase inclusions is presented in Supplementary Fig. S1d, e.

– Minerals occurring in the Matysken Cr-spinel-hosted inclusions are rare, but still present in the host dunite (diopside, pargasite, phlogopite, serpentine, chlorite, garnet), while some minerals in the isoferroplatinum-hosted inclusions are absent in the dunite and never found in association with forsteritic olivine (e.g. albite, K-feldspar, muscovite, quartz) (Fig. 11). Clinopyroxene, namely diopside, is the most abundant mineral in both Cr-spinel- and isoferroplatinum-hosted inclusions. However, its composition plotted together with the clinopyroxene compositions of the Matysken complex major units (dunite, wehrlite, clinopyroxenite) shows that Cr-spinel-hosted diopside is analogous to those from the host dunite, while diopside from isoferroplatinum is enriched in Fe and Al, and does not intercept with the

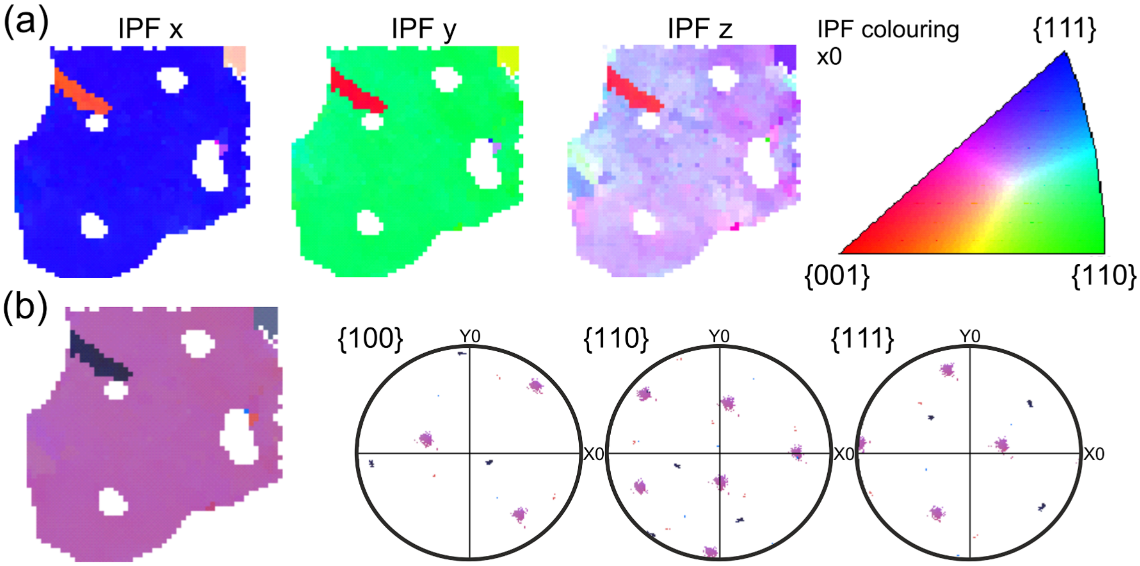

Fig. 9. EBSD data on isoferroplatinum grain from Fig. 8a: (a) – IPF colouring, are overpainted by black is Cr-spinel, (b) – Euler colouring with inversed pole figures.

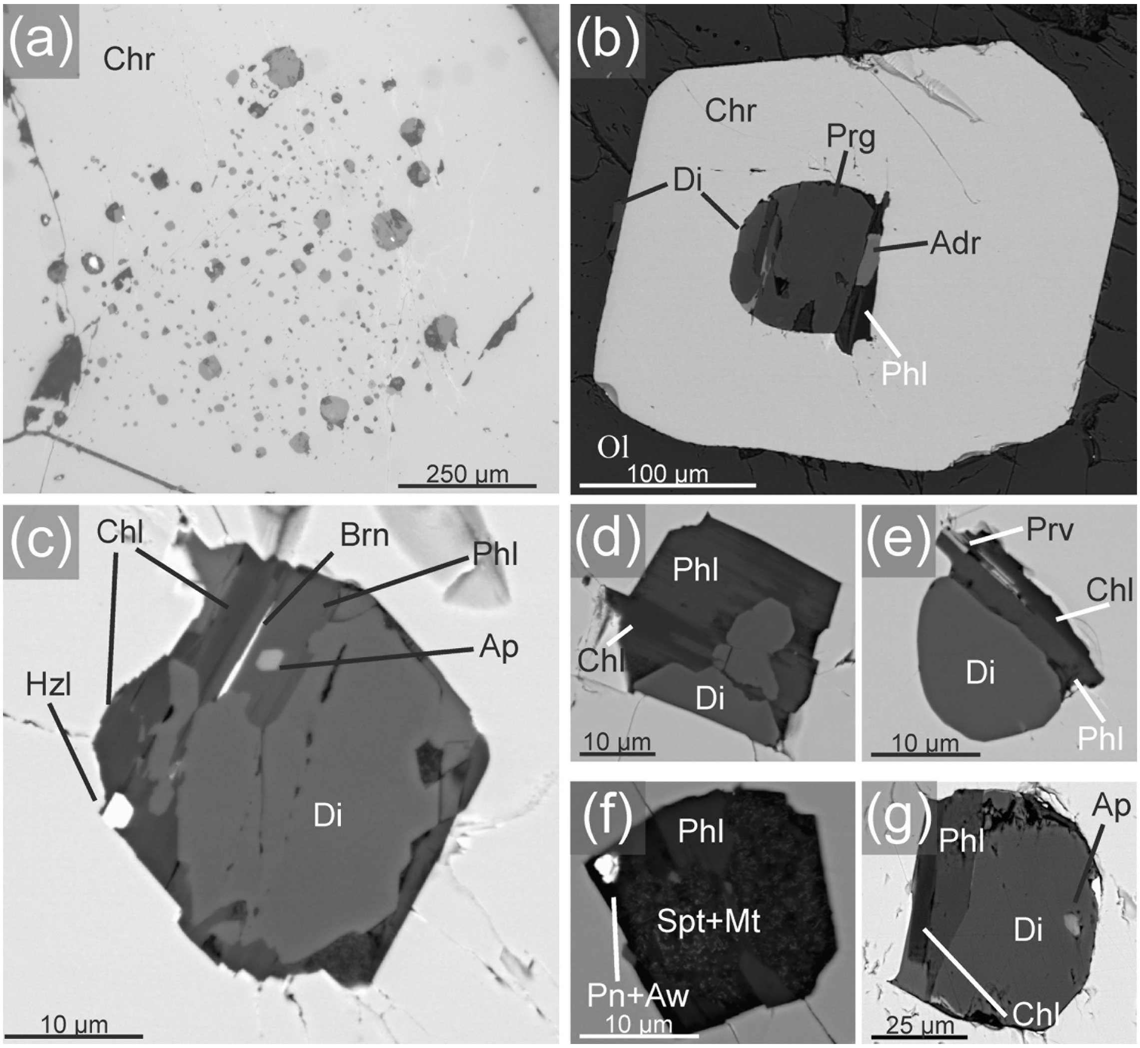

Fig. 10. Multiphase inclusions in Cr-spinel: (a) – general view of a Cr-spinel grain from massif chromitite populated by inclusions, (b) single Cr-spinel grain with an inclusion, (c-g) – inclusions in the Cr-spinel from massif chromitite. (a) – reflected light, all other – BSE. See Supplementary Fig. S2 for more data on Cr-spinel-hosted inclusions.

field of dunitic clinopyroxene (Fig. 12). This is in agreement with observations by Nixon et al. (1990). These authors showed that clinopyroxene in isoferroplatinum-hosted inclusions are more ferric than clinopyroxene from Cr-spinel (Fig. 12):

– Cr-spinel-hosted inclusions are enriched in minerals such as serpentine, chlorite and garnet, which are typical in altered peridotites, while in isoferroplatinum-hosted inclusions these minerals are absent or exceptionally rare (Fig. 11).

At this point, we face an evident paradox, which is that there are compositionally different multiphase inclusions in coexisting Cr-spinel and isoferroplatinum, which presumably formed together. A possible solution involves the role of isoferroplatinum as a protective shell. In this case, inclusions in isoferroplatinum may provide more reliable evidence of mineralizing processes.

Table 1
Comparative review of multiphase inclusions in isoferroplatinum previous studies.

		Cpx	Amph	Pl	Fsp	Ap	Qz	Ep	Tit	Msc	Phl/Bt	Chl	Spt	Tlc	Gl
1	Matysken complex (lodes, present study)	+	+	+	+	+	+	–	+	+	+	+	+	–	–
2	Tulameene complex in British Columbia (placer)	+	+	+	–	–	–	+	–	–	+	+	+	–	–
3	Different placers of British Columbia	+	+	–	–	–	+	–	–	–	–	–	–	+	–
4	Kondyor (placer)	+	+	+*	+*	+	+*	–	–	–	+	+	+	–	–
5	Nizhnytagilsky complex in Urals (placer)	+	+	–	–	+	+	–	+	+	+	+	+	–	+
6	Itchayvayamsky complex in Koryak Highlands (placer)	–	–	+	+	+	+	+	–	–	+	–	–	–	–
7	Ophiolitic peridotites of Koryak Highlands (placer)	+	+	+*	+*	–	+*	–	–	–	+	+	+	–	–
8	Galmoenansky complex in Koryak Highlands (lode)	+	–	–	–	–	–	–	–	–	+	–	–	–	–

Note: 1 – present study, 2 – (Nixon et al., 1990), 3 – (Barkov et al., 2005), 4 – (Mochalov, 2002; Shcheka et al., 2004); 5 – (Cabri and Genkin, 1991; Johan, 2006), 6 – (Sidorov et al., 2019), 7 – (Dmitrenko and Mochalov, 1989; Mochalov, 2002); 8 – (Nazimova et al., 2011). * – minerals were taken as mechanically incorporated during transport in river by Mochalov (2002). Mineral abbreviations: Cpx – clinopyroxene, Amph – amphibole, Pl – plagioclase, Fsp – K-feldspar, Ap – apatite, Qz – quartz, Ep – epidote, Tit – titanite, Msc – muscovite, Chl – chlorite, Spt – serpentine, Tlc – talc, Gl – glass.

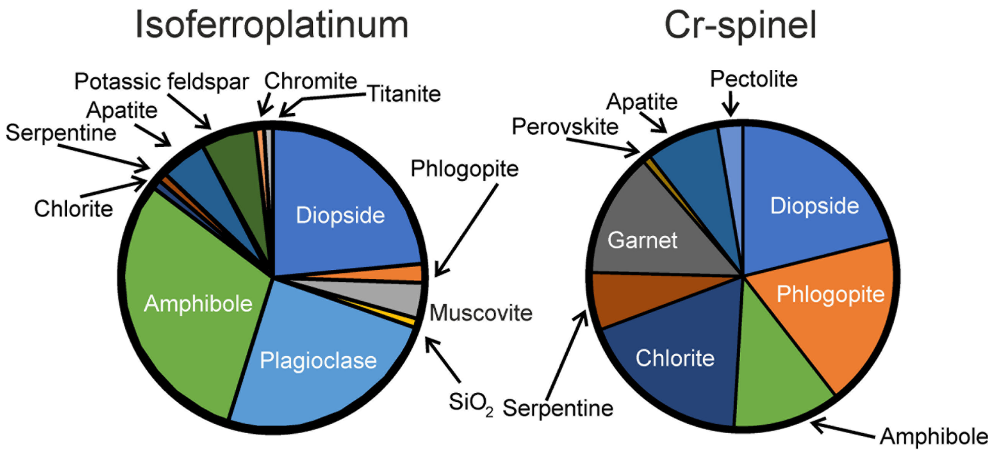

Fig. 11. Relative mineral abundances in the multiphase inclusions in isoferroplatinum (N = 45) and Cr-spinel (N = 50), based on the mineral counts.

5.3. *Origin of platiniferous chromitites: a review*

Several features of platiniferous chromitites should be taken into consideration when constraining the origin of isoferroplatinum-hosted inclusions:

(1) Chromitites and isoferroplatinum occur in close association (Fig. 5). Despite the fact that PGM in Ural-Alaskan type dunite or clinopyroxenite can occur away from Cr-spinel accumulations (e.g. Mochalov and Bortnikov, 2008; Tolstykh et al., 2011), the amount of non-chromitite-related PGM is negligible, hence the processes forming both Cr-spinel and platinum mineralization must be closely related;
(2) Despite the aforementioned affinity between isoferroplatinum and chromitites, the vast majority of the latter lack any PGM with sizes exceeding the first µm. In other words, the distribution of isoferroplatinum between different chromitite bodies is extremely uneven;
(3) Chromitites are podiform, i.e. they are of variable shape, tend to form short schlieren (Fig. 4a, b), which may wedge out and bifurcate in different directions, and strongly differs in shape from chromitites in layered intrusions. This suggests it is unlikely that they have an early-magmatic (cumulate) origin, which is in agreement with previous studies of both Ural-Alaskan type (Augé et al., 2005; Pushkarev et al., 2007; Stepanov et al., 2017) and ophiolitic peridotites (Arai, 1997; González-Jiménez et al., 2014);
(4) The role of sulfides is very minor, despite the fact they are present in most samples. The volume of sulfides is negligible compared to that of isoferroplatinum (Figs. 4 and 7). Subsequently, the partitioning of PGE in a hypothetical sulfide liquid may not be considered to be the main process responsible for PGE enrichment in podiform chromitites.

Among the published work on isoferroplatinum-hosted inclusions, the only attempt to explain their origin in magmatic terms was made by Nixon et al. who proposed that they are the result of "…*silicate melt trapped within platinum alloy at the time of chromitite formation*" (Nixon et al., 1990, page 531). However, this interpretation involves highly evolved Si- and alkali-enriched melt compositions, which were not recorded within the host dunite or its main constituents (i.e. high-Mg olivine and Cr-spinel). In addition, this model stipulates that the differences in the composition of Cr-spinel- and isoferroplatinum-hosted inclusions should be explained as a result of their crystallization from different types of melt.

Two models that attempted to link the origin of large PGM nuggets to magmatic processes are listed below. Augé et al. (2005) suggested that Cr-spinel and PGM crystallized in a magmatic reservoir inside a dunite body: "*An active regime with continued magma injection along conduits (dykes) within the consolidating or consolidated part of the chamber (dunite) could give rise to cavities that would be constantly fed by the magma traversing the dunite body. In this model, chromite, together with minor olivine, would then accumulate within the cavities and thus would be intrusive into the main dunite body…*" (Augé et al., 2005, page 726). However, this hypothesis still does not explain the extreme enrichment of PGE in a given volume of melt, which is crucial to the formation of nuggets. These authors concurred and stated that: *"Although this model can explain the occurrence of both Pt-rich and Pt-poor chromitite, it does not account for the strong and unusual Pt enrichment, suggesting that in Alaskan-type complexes, the accumulation of Pt-PGM by the chromite crystals was very efficient"* (Augé et al., 2005, page 730). To explain this,

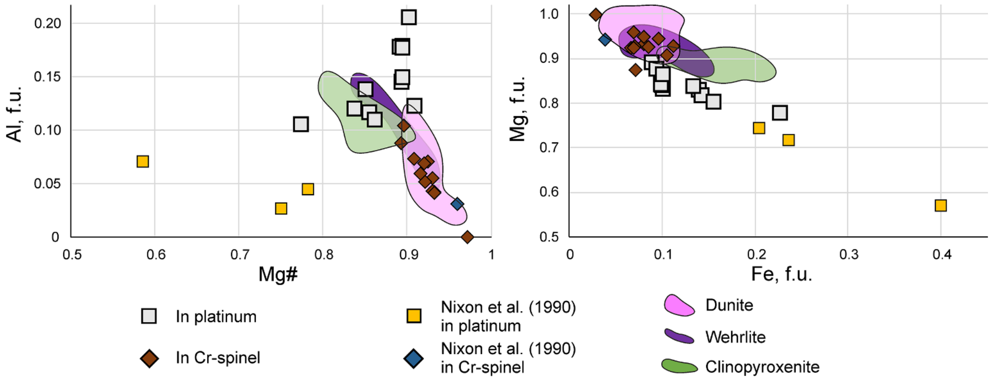

Fig. 12. Composition of clinopyroxenes from multiphase inclusions in isoferroplatinum and Cr-spinel compared with those in ultramafic rocks of the Matysken complex and the inclusions from nuggets related with Tulameene complex (Nixon et al., 1990). See Supplementary Table S3 for compositions.

these authors used the concept of PGM precipitation with chromite, based on local changes in fO_2 caused by chromite crystallization, as proposed by Mungall (2002), in combination with their own idea of prolonged interaction between gravitational concentrations of chromite and fresh magma passing through the cavities (Augé et al., 2005). This may appear to be a satisfactory explanation, but is not supported by our results, because the composition of the inclusions hosted by isoferroplatinum (Fig. 11) has no affinities with mafic/ultramafic melts that can be parental to Cr-spinel. Instead, the recorded feldspar, mica, albite and quartz in multiphase inclusions strongly imply that their parental melt media should be in disequilibrium with the host dunite. Furthermore, according to this model, the same grade of platinum mineralization should be expected for chromitites of approximately the same size. However, this contradicts our results and data from previously published works (Nazimova et al., 2011).

According to the second model, podiform chromitites are the result of two consecutive immiscibility events: (1) the partitioning of ultramafic silicate melt into silicate and Cr-rich oxide liquid, and (2) the subsequent partitioning of the latter into a Cr-rich liquid and molten metal (Okrugin, 2011). The intergrowths of PGM and hydrous silicates are assumed to reflect the partitioning of incompatible elements into liquid metal. Although this model may explain the existence of large isoferroplatinum crystals and some textural features, such as isoferroplatinum cementing euhedral Cr-spinel and intergrowths between isoferroplatinum and hydrous silicates, it has two obvious limitations. Firstly, there is a lack of any published experimental data which may support such immiscibility and existence of both Cr-rich oxide and Pt-Fe liquids at crustal magmatic temperatures. Ideas on silicate-chromite liquids immiscibility have been long before proposed for the origin of chromite seams in the Bushveld complex (Mcdonald, 1965), but were subsequently rejected as unrealistic (Jackson, 1966). The second controversy is the lack of any textural evidence of Cr-rich oxide liquid existing in the Matysken complex, i.e. no droplet-shaped Cr-spinel grains were observed in dunites.

Some of the above mentioned problems may be circumvented if the late-magmatic crystallization of the Cr-spinel was assumed to be caused by a hydrous boninitic melt, which infiltrated through ultramafic rocks and reacted with them, followed by the dissolution of pyroxene and the subsequent formation of chromitite-bearing dunite in ophiolites (Arai, 1997). This model, which is at the interface of magmatic and post-magmatic processes, does not contradict aforementioned constraints on the origin of platiniferous chromitites. Instead, it provides insights into some textural features of different chromitite occurrences in dunites along with the peculiar composition of Cr-spinel-hosted inclusions. However, a large melt volume would be required to have circulated through the system and thus created a wide halo of hydrous and SiO_2-, K- and Na-rich minerals around PGE-rich chromitite veins, which are absent in the studied samples. Moreover, this hypothesis is not applicable to unusual compositions of isoferroplatinum-hosted inclusions.

5.4. Alternative non-magmatic concepts for the origin of platiniferous chromitites

We propose that the presented data on isoferroplatinum-hosted inclusions are in disagreement with a magmatic origin for the formation of platiniferous chromitites. An alternative explanation for PGM and Cr-spinel paragenesis may potentially involve non-magmatic mechanisms of Pt accumulation together with Cr-spinel. The only attempt to explain isoferroplatinum-hosted inclusions in non-magmatic terms was made by Mochalov (2002), who assigned the origin of such minerals like quartz, albite and K-feldspar to mechanical entrapment in a placer, but then explained that mica and amphibole inclusions in isoferroplatinum resulted from post-magmatic fluid-assisted processes. Unfortunately, this proposal lacks any detail which can assist in uncovering the exact mechanism of PGM formation. However, such ideas are consistent with the reports of PGM occurrences in re-crystallized parts of dunite units in Ural-Alaskan type complexes (Mochalov, 2013; Nazimova et al., 2011; Pushkarev et al., 2007). The above mentioned arguments for simultaneous crystallization of Cr-spinel and isoferroplatinum, as well as general similarities of multiphase inclusions in these minerals, demands the application of potential non-magmatic models for the formation of Cr-spinel and isoferroplatinum, and their entrapped inclusions.

One possible explanation is based on a study of chromitites from the Nizhnytagilsky complex, where Cr-spinel-hosted inclusions of silicates and silicate-PGM intergrowths are attributed to crystallization from a post-magmatic liquid, which was extracted from interstitial assemblage of dunite during the latest stage of rock deformation (Pushkarev and Anikina, 2002; Pushkarev et al., 2007). This liquid was enriched in Ca, alkalis and saturated with extremely fine (μm-scale) particles of Cr-spinel. According to this model, the aggregate of Cr-spinel, silicates and PGM precipitated from this liquid later recrystallized, resulting in chromitites with inclusions of silicates and PGM.

Although this concept seems speculative, it avoids numerous problems encountered in the formation of chromite- and isoferroplatinum-hosted inclusions, such as the contrast between inclusion constituents and host dunite, and thus warrants further investigation. The idea of platiniferous chromitites being the result of recrystallization is also supported by our data. For example, this concept can explain equigranular mosaic textures of chromitites with straight boundaries and 120° triple junctions (Fig. 5c), which are the result of mineral recrystallization (Pike and Schwarzman, 1977) and the discordancy between the inclusion borders and crystallographic axis of host isoferroplatinum (Fig. 9).

Another problem is that any model attempting to explain platiniferous chromites on the basis of recrystallization automatically involves the notion of dunite being a modified rock, rather than primary magmatic product (cumulate). This may appear bold, but similar ideas have also been proposed for ultramafic restites in complexes of other types and have since gained acceptance. For example, the origin of replacive dunite during the metasomatism of harzburgite in ophiolites is widely accepted (Melcher et al., 1997; Su et al., 2016 and references therein). The idea of prolonged subsolidus evolution of dunite in Ural-Alaskan type complexes has also been proposed (Mochalov, 2013; Nazimova et al., 2011). The view that Cr-spinel-PGM mineralization in ophiolites occurred due to multiple hydrous alteration/recrystallization events was advocated for the formation of Cr-spinel-hosted multiphase inclusions (Borisova et al., 2012; Gervilla et al., 2012; González-Jiménez et al., 2014). Similar ideas, involving the dissolution and subsequent reprecipitation of Cr-spinel, were proposed in several other publications (Arai and Akizawa, 2014; Pushkarev et al., 2015).

The most advanced sense of understanding the nature and origin of isoferroplatinum-hosted inclusions involves multiple events of post-magmatic alteration and subsequent recrystallization of chromitites that result in the formation of chromite-isoferroplatinum aggregates: "*…indistinguishable from the original euhedral magmatic platinum-group minerals — at least in terms of their microstructures and major-element compositions*" (González-Jiménez et al., 2014). Taking this into consideration, inclusions in isoferroplatinum and Cr-spinel can be explained as a mixture of precursor minerals, which were modified by fluid-assisted processes coeval with recrystallization of isoferroplatinum and Cr-spinel. During this process, isoferroplatinum armored minerals from interacting with host high-Mg olivine, which explains the presence of minerals such as K-feldspar, albite or silica in the inclusions within isoferroplatinum-hosted and their absence in Cr-spinel-hosted inclusions.

6. Conclusions and pending problems

(1) A primary origin for isoferroplatinum-hosted multiphase inclusions from lode chromitites is supported by: (a) absence of any signs of isoferroplatinum alteration along the contacts with inclusions, (b) the low alteration level of the dunite, and (c) the absence of

fractures transecting inclusions, which could be interpreted as potential pathways for infiltrating fluids, point towards the primary nature of isoferroplatinum-hosted multiphase inclusions. Thus inclusions in isoferroplatinum and Cr-spinels are an essential tool for understanding the origin of platinum mineralization.

(2) Inclusions in isoferroplatinum and Cr-spinel can occur within a single sample and share some common features such as shape and mineralogical contrast with the host dunite mineral assemblage, thereby pointing towards a similar process of formation. This idea is supported by the close spatial association and textural relationship of isoferroplatinum and Cr-spinel. Our study demonstrates that none of the minerals within inclusions can be considered undoubtedly euhedral and thus neither formed earlier than the other. In addition, there are strong compositional differences between inclusions in Cr-spinel and isoferroplatinum, where the latter are more felsic. This may be explained by isoferroplatinum forming an impermeable barrier, which allowed for better preservation of the initial mineral assemblages within the inclusions from interaction with the host dunite.

(3) The unusual composition of inclusions in isoferroplatinum places contraints on the idea that isoferroplatinum precipitated from ultramafic, basaltic or compositionally similar silicate melts. Existing genetic models advocating intrinsic magmatic processes should be further scrutinized and critically reevaluated using newly obtained data on multiphase inclusions hosted in coexisting isoferroplatinum and Cr-spinel.

(4) An alternative concept which may help to understand both the origin of platiniferous chromitites and inclusions in isoferroplatinum involves the subsequent hydration and recrystallization of the host dunite or what its precursor may have experienced during the post-magmatic stage. The role of recrystallization processes is also supported by equigranular mosaic textures of chromitites with 120° angles between Cr-spinel crystals. However, a complete model of this process is still under development.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.oregeorev.2020.103367.

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