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Role of volatiles in intrusion emplacement and sulfide deposition in the supergiant Norilsk-Talnakh Ni-Cu-PGE ore deposits

Stephen J. Barnes¹,*, Marina A. Yudovskaya^{2,3}, Giada Iacono-Marziano⁴, Margaux Le Vaillant¹, Louise E. Schoneveld¹, and Alexander R. Cruden⁵

ABSTRACT

The Norilsk-Talnakh orebodies in Siberia are some of the largest examples on Earth of magmatic Ni–Cu–platinum group element (PGE) deposits, formed by segregation of immiscible sulfide melts from silicate magmas. They show distinctive features attributable to degassing of a magmatic vapor phase during ore formation, including: vesiculation of the host intrusions, widespread intrusion breccias, and extensive hydrofracturing, skarns, and metasomatic replacement in the country rocks. Much of the magmatic sulfide was generated by assimilation of anhydrite and carbonaceous material, leading to injection of a suspension of fine sulfide droplets attached to gas bubbles into propagating tube-like host sills ("chonoliths"). Catastrophic vapor phase exsolution associated with a drop in magma overpressure at the transition from vertical to horizontal magma flow enabled explosive propagation of chonoliths, rapid "harvesting" and gravity deposition of the characteristic coarse sulfide globules that form much of the ore, and extensive magmatic fluid interaction with country rocks.

INTRODUCTION

The Ni–Cu–platinum group element (PGE) deposits of the Norilsk-Talnakh camp in Arctic Siberia are among the most valuable economic metal accumulations of any kind on Earth (Barnes et al., 2020). They are part of the Siberian large igneous province, which was cataclysmically emplaced ca. 252 Ma, synchronous with the Permian-Triassic mass extinction (Kamo et al., 2003; Burgess and Bowring, 2015). The deposits have been extensively studied for more than a century, yet many aspects of their genesis remain poorly understood.

The currently dominant paradigm for these and other magmatic sulfide deposits of this type is the "conduit model" (Rad'ko, 1991; Naldrett et al., 1992), whereby ore formation is inferred

to take place in subvolcanic feeder conduits as a result of assimilation of crustal sulfur (S) by silicate magma, formation of entrained immiscible sulfide liquid, scavenging of Ni, Cu, and PGEs by that liquid, and final deposition in mechanical trap sites. Since the formulation of this model, numerous lines of evidence have emerged to challenge key aspects, particularly

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at Norilsk. Detailed local geologic data from Russian sources have become available (reviewed by Barnes et al., 2020) that are inconsistent with the interpretation of the Norilsk intrusions as feeder conduits to the overlying lavas. Important advances have been made in understanding the physics of sulfide liquid droplets (Robertson et al., 2015; Yao et al., 2020) and the role of magma degassing and vapor-related transport and deposition of sulfide liquids in ore formation (Mungall et al., 2015; Iacono-Marziano et al., 2022). Analogue experiments have provided new insights into the mechanisms of dike and sill propagation (Kavanagh et al., 2015). Here, we summarize these developments and synthesize them into a new model for the explosive emplacement of the host intrusions and sulfide ores, accounting for hitherto neglected magmatic-hydrothermal characteristics.

NORILSK-TALNAKH GEOLOGY

The Norilsk-Talnakh region (Fig. 1) contains a roughly 5-km-thick stack of tholeiitic basalts, forming part of the vast ca. 250 Ma Permian—Triassic Siberian large igneous province (SLIP). The ore deposits are found within a very specific intrusive component of the SLIP: a suite of shallow, strongly differentiated mafic-ultramafic intrusions that intrude the Carboniferous to Upper Permian platform sequence, which is more than 3 km thick, comprising clastic sediments, carbonates, evaporites (containing anhydrite and abundant bituminous material), and carbonaceous rocks (argillites and coal). The Talnakh and Kharaelakh intrusions that host the major deposits are emplaced at two distinct levels within the Devonian and Permian sediments, respectively (see the Supplemental Material1), <2 km below the Paleozoic land surface. These are unusually shallow depths for this class of ore deposit, compared with typical depth ranges of 3–25 km (Naldrett, 2004).

The ore-bearing intrusions are roughly skishaped or wing-shaped elongate sills, representing a type of mafic-ultramafic intrusion called chonoliths (Fig. 1). They are dominantly gabbroic, with olivine-rich cumulates toward the base and highly contaminated inclusion-rich upper and lower margins. Mine-scale geology has shown that some of these bodies, particularly the Kharaelakh intrusion, are closed off at the propagating "frontal" end (Krivolutskaya et al., 2018). Host-rock lithology played a key role in localizing magma emplacement at particular horizons (e.g., Godlevsky, 1959; Naldrett, 2004), involving both injection into and replacement of the host sediment.

The origin of such tube-shaped intrusions remains an unsolved problem. Several models have been proposed, mostly involving development of finger-like lobes at the propagating edges of sills and dikes (Magee et al., 2016; Köpping et al., 2023), but based on the evidence of extensive mine-scale drilling, the Norilsk-Talnakh bodies cannot be interpreted as the leading edges of extensive sheet-like intrusions.

CRITICAL OBSERVATIONS AND CONSTRAINTS ON GENETIC MODELS Interaction with Country Rocks

Based on isotopic evidence, the NorilskTalnakh orebodies are widely thought to have derived their S from assimilation by basaltic melt of the anhydrite-rich sedimentary sequence (Gorbachev and Grinenko, 1973; Grinenko, 1985). Naldrett (2004) suggested that coupled assimilation of coal or other carbonaceous material provided the reductant essential to produce immiscible sulfide melt from dissolved sulfate in the silicate magma.

Potential reductants are carbonaceous shales along with abundant bitumen and other hydrocarbons within the Devonian evaporites and carbonates (Ryabov et al., 2018). Iacono-Marziano et al. (2017) demonstrated this mechanism experimentally and showed that this process simultaneously generates C-H-S-bearing fluids in close physical association with fine sulfide liquid droplets.

Semi-massive sulfide veins and disseminated ores developed within flanking and overlying sedimentary rocks, and also beneath the intrusions, above or below the underlying massive sulfide orebodies (Lightfoot and Zotov, 2014). These Cu-rich "exo-contact" assemblages are referred to as cuprous ores. Their wide variety of textures shows clear evidence of dynamic intrusion involving mechanical injection, impregnation, partial melting, and chemical dissolution of sedimentary host rocks.

The major deposits are flanked by a variety of distinctive skarns and hornfelses showing extensive brecciation, with later salt-anhydritecarbonate veining and selective replacement of fine-scale bedding by sulfide within marl and argillite (Fig. 2). These features developed within extensive hydrothermal alteration aureoles extending $\sim\!200$ m away from mineralized intrusions and only a few meters around barren ones (Pokrovskii et al., 2005). Country rocks were evidently invaded extensively by overpressured magmatic fluids.

Evidence for Vapor-Phase Saturation within the Intrusions

Several lines of evidence indicate that the magmas were volatile-saturated at the time of ore deposition. Infilled vesicles are commonly attached to sulfide globules in the distinctive globular ores that account for much of the sulfide budget in the intrusions. The vesicles take the form of subspherical gaps within the cumulus framework of these olivine orthocumulate rocks (Figs. 3A–3C); they are filled with the crystallization products of strongly differentiated trapped intercumulus silicate liquid, and variable components of hydrothermal minerals, forming "segregation vesicles" and amygdules (Barnes et al., 2019). These features are interpreted as sulfide liquid droplets that were physically attached to gas bubbles, a phenomenon recognized in high-temperature experiments and attributed to the mutual surface affinity of the two phases (Mungall et al., 2015). Crystallization under supercooled conditions (Donaldson, 1976) is indicated by the minor but widespread presence of skeletal olivine grains in picrodolerite (Fig. 3D).

A similar but more complex association occurs in the low-S PGE ores toward the upper margins of the intrusions (Sluzhenikin et al., 2020). These ores contain Pt and Pd in concentrations of a few parts per million, similar to the globular ores below, but with much less sulfide. They are characterized by abundant vesicles, typically with a higher proportion of lower-temperature amygdule filling to segregated silicate melt, coating of bubbles by oxide films, and the presence of irregular mats or schlieren of Tichromite and Cr-magnetite (Fig. 3C; Chayka et al., 2020; Schoneveld et al., 2020; Sluzhenikin et al., 2020; Chayka et al., 2023). The proportion

of bubbles to sulfide, and the proportion of platinum group minerals (PGMs) to sulfides, is much greater than that in the globular ores. In contrast to the globular ores, where the PGMs are spatially attached to sulfides, PGMs in the low-S ores are mainly isolated, separated at a scale of millimeters from the sulfide droplets, which show evidence of having lost S to an escaping volatile phase (Barnes et al., 2021; Gritsenko et al., 2022), a process

that stabilizes PGMs and also raises the effective PGE concentration of the remaining sulfide (Peregoedova et al., 2004; Iacono-Marziano et al., 2022).

Mass Balance and Timing of Sulfide Liquid Formation

The orebodies present a mass balance problem: The aggregate mass of chalcophile elements, particularly Pt and Pd, is much greater than could have been derived from a volume of magma comparable to that of the host intrusions. Naldrett et al. (1992) estimated that a volume of magma 15-200 times the volume of the intrusions would have been required to supply the PGE. A more conservative estimate based on known deposit dimensions, an estimate of 2.5 wt% sulfide in the Kharaelakh intrusion (Zen'ko and Czamanske, 1992), and an estimate of 20 ppb Pt + Pd in the parent silicate magma (maximum contents measured in lavas of the Noril'sk region; Naldrett, 2004) gives a lower estimate of around 10 times the volume of the intrusion, still requiring a large excess. These constraints rule out models that involve in situ sulfide liquid generation entirely within the intrusions themselves. Hence, most of the generation of and metal scavenging by the sulfide liquid must have taken place prior to transport of sulfide droplets into the sills. This requires that assimilation of the anhydrite and the C-rich reductant must have happened either in some deepseated subchamber, as modeled by Yao and Mungall (2021), or within a feeder sill-dike complex, involving a total volume of magma of at least 100 km3 based on the PGE mass balance argument. Magma flow in this system may have been as much lateral as vertical, based on evidence for lateral injection of large dikes in large igneous provinces (Ernst et al., 1995), such that an unseen "deep chamber" may not have been necessary.

Droplet Physics, Bubble Formation, and Sulfide Harvesting

The large size of the sulfide droplets in the globular ores (Fig. 3) presents a problem. If droplets were indeed transported over kilometer-scale vertical distances (Yao and Mungall, 2021), they must have been of the order of hundreds of microns or at most a few millimeters in size; larger droplets would either have settled out or broken up due to shear stress in the emulsion (Robertson et al., 2015). The >2 cm droplets observed in the disseminated ores (Fig. 3A) can only have been generated very close to the site of deposition. Analogue experiments (de Bremond d'Ars et al., 2001) and theory (Robertson et al., 2015) predict that coalescence of fine sulfide droplets is unlikely during transport in dilute emulsions. Experiments by Iacono-Marziano et al. (2022) showed that generation of vapor bubbles during or after generation of sulfide liquid droplets causes multiple small droplets to become collected on the surfaces of the bubbles, thus providing a means by which fine droplets will rapidly coalesce into large ones. This requires that a burst of bubble nucleation and droplet "harvesting" took place synchronous with deposition. Two distinct episodes of vapor bubble generation may have occurred: the first associated with assimilation and generation of a population of small transportable droplets, and the second coincident with intrusion emplacement.

Sill Inception and Overpressure Release

Laboratory experiments by Kavanagh et al. (2015) using gelatine and water as analogues for upper-crustal rocks and mafic magma, respectively, found that a large transient release of

overpressure occurs when a vertically ascending dike transitions to a horizontally propagating and vertically inflating sill. The associated stress drop scales to equivalent values in nature from $\sim\!250$ to 100 MPa at depths between 1 and 10 km. In cases where the magma is already at volatile saturation, this pressure drop would be accompanied by a sudden catastrophic degassing event, causing rupturing of country rocks and rapid injection of magma into the sill. Kavanagh et al. (2015) proposed this pressure drop as a potential trigger for volcanic eruptions, but it may also explain the sudden pressure drop associated with the initial emplacement of the host chonoliths, with important consequences, including:

- 1. Conversion of transported fine droplet "mist" into a "rain" of large globules due to harvesting by a new generation of vapor bubbles;
- 2. Wall-rock fragmentation, creating fracture networks for magma injection, accompanied by intrusion breccias and highly contaminated marginal zones;
- 3. Extensive volatile influx on scales of 10–100 m into the reactive country rocks, giving rise to hydrofracturing, breccias, and skarns; and
- 4. Generation of "damage zone" footwall fracture arrays, facilitating invasion by sulfide liquid to form exo-contact massive ores, localized degassing at the leading edge of the sill, causing pore-fluid overpressure, and channeling of silicate magma into resulting fractures to form the chonoliths.

NEW EMPLACEMENT MODEL

A vertically propagating narrow dike (Fig. 4A) channels a volatile-saturated, overpressured magma containing a fine mist of sulfide droplets rafted on vapor bubbles previously generated during assimilation within the magma column. On reaching a horizon of weakness or competency contrast (e.g., the argillite-bearing Razvedochninsky formation for the Kharaelakh intrusion and the coal-bearing Tunguska group for the Talnakh and Norilsk 1 intrusions; see the Supplemental Material), a new sill begins to propagate as a chonolith, resulting in a sudden drop in overpressure and collapse of the dike (Fig. 4C).

The droplet-harvesting process generates a wide range of bubble to droplet ratios (Fig. 4B), from bubbles with very little sulfide to virtually bubble-free sulfide that detaches from the vapor phase under its own weight (Yao et al., 2020). Dense sulfide-rich droplet-bubble pairs sink rapidly; the earliest sinking globules reach the floor and coalesce to form the basal massive ores, whereas later ones accumulate into the accreting bed of olivine cumulates. Bubbles with little or no attached sulfide float to the roof, becoming coated with spinel grains and accumulating in the upper taxite breccias. Ongoing degassing here drives destruction of sulfide, growth of PGMs, and the formation of the low-S PGE zone. A continuing influx of magma splits the two ore zones (S rich and S poor) and eventually crystallizes to form the relatively uncontaminated olivine-poor central gabbro-dolerites (Fig. 4D).

This model is by no means the first to incorporate volatiles (e.g., Marakushev et al., 2003), but it is the first to link explosive intrusion emplacement with a mechanism for collecting a large volume of previously generated dispersed sulfide droplets. While it leaves some complex problems unaddressed, such as the mismatch between the Pt/Pd ratios of the orebodies and the likely parent basaltic magmas, extensive differentiation of the sulfide liquid, and the nature or even existence of staging magma chambers, it provides a testable working hypothesis.

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Figure captions

Figure 1. (A) Location map. (B–C) Long section through Kharaelakh (K) and Talnakh (T) intrusions. (D) Schematic three-dimensional view of a "composite" Norilsk-Talnakh chonolith complex, showing internal differentiation, disposition of ore types, and flanking sills and breccias.

Figure 2. Skarn and breccia ores in Talnakh intrusion footwall. (A) Footwall below massive sulfide in Talnakh intrusion: Sandstone blocks (darker) are boudinaged among microfolded mudstone (lighter); Skalisty Mine. (B) Sulfide-rich bands in banded forsterite-monticellite skarn after marl; serp—serpentinite; f-grained—fine-grained. (C) Massive sulfides and veinlets rimmed by serpentinized forsterite skarn (FS) in brecciated marble. (D) Banded massive sulfides along relict bedding in meta-argillite hornfels.

Figure 3. Globular ores. (A) Large sulfide globules in taxitic picrodolerite (PGD). (B) Phase map of PGD (from synchrotron X-ray fluorescence microscopy [XFM] image) showing sulfides (sulf) and silicate cap within void in olivine framework. (C) Upper taxite, segregation vesicles, and sulfides within Crspinel mat (Cr-Fe-Ti XFM map). (D) Picrodolerite with skeletal olivine (green) and segregation vesicles (Cr-Fe-Ca XFM map). Minerals: amyg— amygdule, ap—apatite; Ccp—chalcopyrite; cpx—clinopyroxene; hb/h'blende—hornblende; Ol/olv—olivine; plag/plg—plagioclase; Pn—pentlandite; Po—pyrrhotite.

Figure 4. Schematic illustration (not to scale) of model for explosive emplacement and sulfide droplet deposition in propagating chonolith. (A, B) Time T1: Inception of chonolith and explosive injection of volatile-saturated magma containing transported fine sulfide droplets. (C, D) Time T2: Ongoing magma injection and inflation of chonolith. (E) Microcomputed tomography (micro-CT) image of experimental run showing multiple sulfide droplets (yellow) "harvested" by a gas bubble (blue), ~1 mm across. PGE—platinum group element.

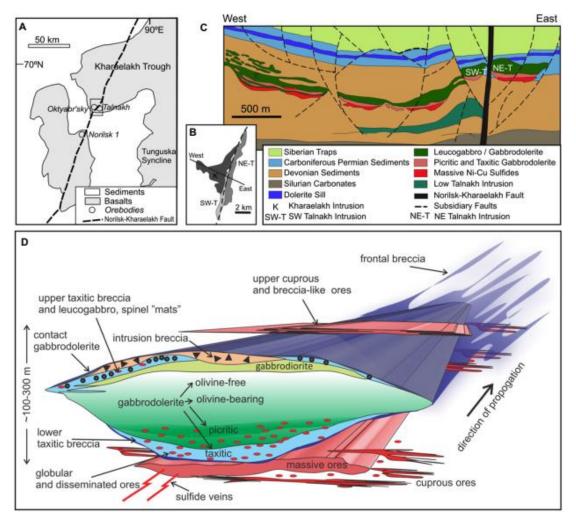


Figure 1

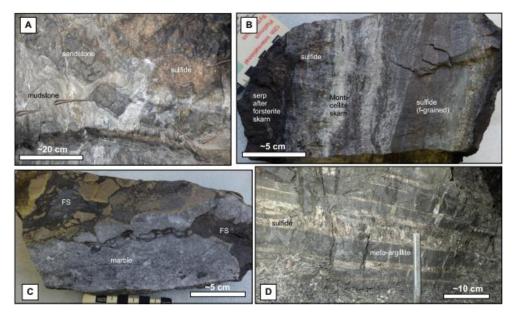


Figure 2

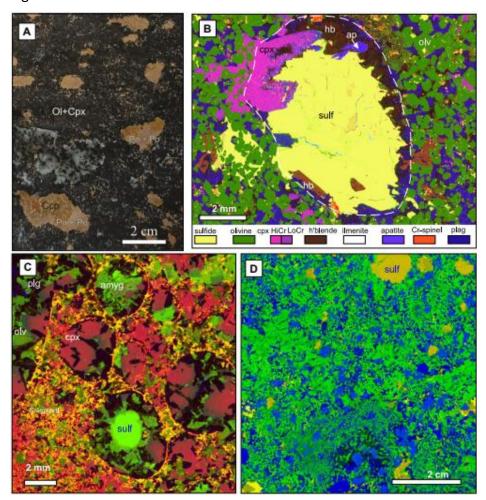


Figure 3

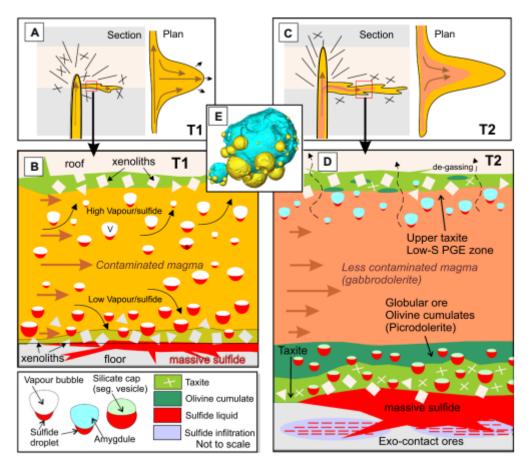


Figure 4