

Origin of zoned laurite – a platinum group mineral from the Mesoarchean Bangur chromite deposit, Boula–Nuasahi ultramafic complex, Odisha, India

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Platinum group minerals (PGMs) belonging to laurite (RuS₂) – erlichmanite (OsS₂) solid solution series and Os–Ir–Ru alloys generally occur as minute inclusions (<10 µm) in both stratiform and podiform chromitite deposits. In a recent study of chromitite samples from Bangur chromite deposit in the Boula–Nuasahi ultramafic complex, Odisha, India, we found a relatively coarse-grained (~30 µm) laurite crystal in addition to other PGMs. This laurite occurs as an included crystal within a ferritchromite grain and has a chemical composition (wt%): Ru ~ 43–45, Os ~ 12–15, S ~ 35, Ir ~ 2–3 and As ~ 0.2, as measured by electron probe micro-analyzer. Due to the high osmium content, it is named as Os-laurite. The mode of occurrence, texture, structure and chemical composition of laurite support a magmatic origin for this mineral that might have crystallized at a temperature slightly lower than 1200°C. The unusual and complex oscillatory zoning (i.e. Os-rich and Os-poor bands) observed within the laurite could have developed below 1200°C (but above 1000°C) in a late magmatic stage due to local fluctuations in the surrounding thermodynamic conditions, primarily the sulphur fugacity.

Keywords: Chromitite, laurite, platinum group minerals, magmatic deposit, osmium, ultramafic rocks.

ECONOMICALLY extractable platinum group mineral (PGM) deposits in the earth's crust are generally sparse, although PGMs can be found associated with various mantle rocks such as ophiolites, magmatic base metal sulphides and chromitite bodies¹. In India, the only potential PGM deposit has been located in the Boula–Nuasahi–Bangur area, Odisha². Other occurrences are reported from the Sitampundi complex, Tamil Nadu; Hanumalapura, Karnataka; and Gondpipri area, western Bastar Craton, Maharastra^{3–5}. The Mesoarchean Boula–Nuasahi ultramafic complex (BNUC) occurring in the southeast border of the Singhbhum Craton consists of mainly four litho-units: gabbro-anorthosite, pyroxenite, peridotite that hosts a series of chromitite lodes, and a slightly younger, coarse-grained gabbro unit known as the Bangur gabbro (Figure 1). The Bangur gabbro (~3.1 Ga) has a discordant relationship with these litho-units, and the

intrusion of this gabbro has structurally disturbed the area with large-scale brecciation that formed a long, narrow, tongue-shaped breccia apophysis^{6,7}.

This late magmatic event which is associated with hydrothermal base metal sulphides, has caused extensive hydrothermal alteration to the earlier litho-units and is considered the cause of Pt–Pd mineralization in BNUC^{8,9}. Various mineralization processes and the PGMs of this area have been reasonably well studied with respect to their mineralogy, mode of occurrence, association, economic potential, geochemistry and genesis^{9–16}. Augé *et al.*⁸ have provided a detailed list of PGMs occurring in this ultramafic belt and have recognized three types of platinum group element (PGE) mineralization and PGM assemblages, viz. type-1 (magmatic), type-2A (hydrothermal, associated with base metal sulphides) and type-2B (hydrothermal, without base metal sulphides). Occurrences of laurite (RuS₂) have been reported by them in all the above three types of mineralization⁸. In this area, PGMs generally occur as very fine sized crystals (<10 µm) and very rarely they exceed 15 µm. During our recent studies on some chromitite samples from the Bangur chromite mines, apart from other PGMs (e.g. sudburyite), we identified some laurite grains, of which one was a relatively large (~30 µm) and rare type of laurite crystal showing anomalous zoning. Generally, the IPGE (Ir-group PGE: iridium (Ir), osmium (Os), ruthenium (Ru))-bearing minerals such as laurite–erlichmanite show internal zoning of Os and the compositional variations in laurite–erlichmanite reflect the crystallization conditions of PGMs and their host chromites. Therefore, these can be used as a petrogenetic indicator¹⁷. In the Nuasahi area, Augé *et al.*⁸ mentioned the absence of zoning in laurite crystals in the type-1 assemblage, except for one crystal which showed an Os-rich core, but neither any details were presented, nor any explanation was provided for the same. Zaccarini *et al.*¹⁸ have reported the occurrence of very fine-grained laurite crystals (~5 µm) having erlichmanite rims from the Nuasahi area close to the breccia apophysis. According to Mondal and Zhou¹⁴, all the IPGEs of the sulphide-rich breccia assemblage have originated from ultramafic–chromitite breccia. Therefore, in the present case, the origin of such a large laurite crystal (a sulphide phase) having an irregular zoning pattern within a ferritchromite grain (an oxide phase) needs to be explained. In this communication, we present the characteristics of laurite, discuss its origin and explain the reasons for this type of abnormal zoning pattern.

The zoned laurite crystal was identified in a brecciated ore sample collected from Bangur area that consisted of chromitite clasts engulfed within the Bangur gabbro (Figure 2 a). The laurite crystal was present within a ferritchromite grain which had been partially/locally replaced by hydrothermal chlorite (Figure 2 b). The associated ferritchromite grains had corroded and/or semi-rounded grain boundaries. In the same sample, the fractures and crystallographic planes were also partly replaced by chalcopyrite at some places, which matches the ferritchromites described

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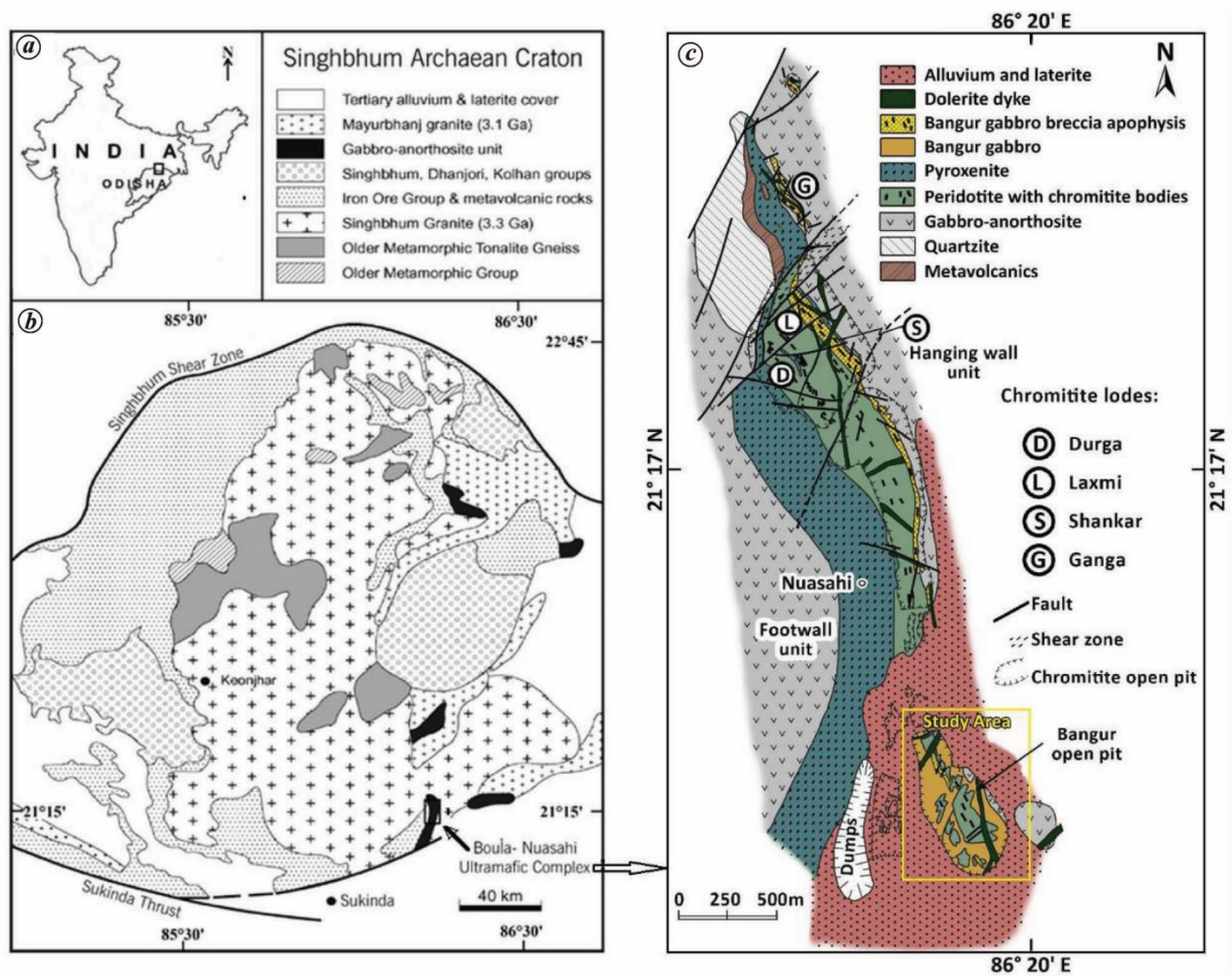


Figure 1. *a*, Location of the Singhbhum Archaean Craton. *b*, Simplified geological map of Singhbhum Archaean Craton showing the location of Boula–Nuasahi ultramafic complex (BNUC) (modified after Saha⁶ and Mondal *et al.*³¹). *c*, Geological map of BNUC showing the Bangur study area (modified after Augé *et al.*⁸, and Augé and Lerouge⁹).

by Augé and Lerouge⁹ from the ‘breccia apophysis’ mapped in the BNUC area. The laurite crystal had an euhedral shape and shared sharp grain boundaries with the ferritchromite grain on three sides (Figure 2 *c*). The laurite grain had the composition (in wt%): Ru ~ 43–45, Os ~ 12–15, S ~ 35, Ir ~ 2–3 and As ~ 0.2 (Table 1), as measured by wavelength dispersive spectrometry (at 25 kV accelerated voltage and 27 nA beam current with 1–2 μm spot size); JEOL JXA-8900R electron probe micro-analyzer (EPMA) at the Institute of Applied Mineralogy and Economic Geology, RWTH Aachen University, Germany. A natural chalcopyrite standard and synthetic alloys of Os, Ru, Ir were used for calibration, and the precision was better than 1%. The chemical composition of laurite revealed Os enrichment, and therefore, it can be named as an Os-laurite. The Os-laurite grain in the study of our sample showed complex oscillatory zoning with Os-enriched bands (Figures 2 *d* and 3). Figure 4 presents the energy dispersive analytical X-ray (EDAX) peaks of Os-laurite and its Os-rich bands.

Laurite (RuS_2)–erlichmanite (OsS_2) solid solution series and Os–Ir–Ru alloys are found in both stratiform and podiform magmatic chromitite deposits, where most of the laurite–erlichmanite grains occur as minute mineral inclusions forming anhedral to euhedral crystals, in single or composite associations with other PGMs, base metal sulphides and/or silicates in chromitites^{16,19–25}. Textural characteristics suggest these laurites be of magmatic origin, which co-precipitate with chromite on the liquidus of basaltic melts^{20,26}. This indicates that a sulphide mineral can crystallize directly from a silicate melt. Based on the single-spot analysis and X-ray elemental mapping using EPMA, many studies have revealed that laurite and erlichmanite that are hosted in chromitites show mutual substitution between Ru and Os^{27–29}. This type of substitution of Ru by Os in laurite and vice versa in erlichmanite produces different zoning patterns within the laurite–erlichmanite grains³⁰, which can be grouped into three main types: (i) normal zoning showing Os-poor core (laurite) and Os-rich rim,

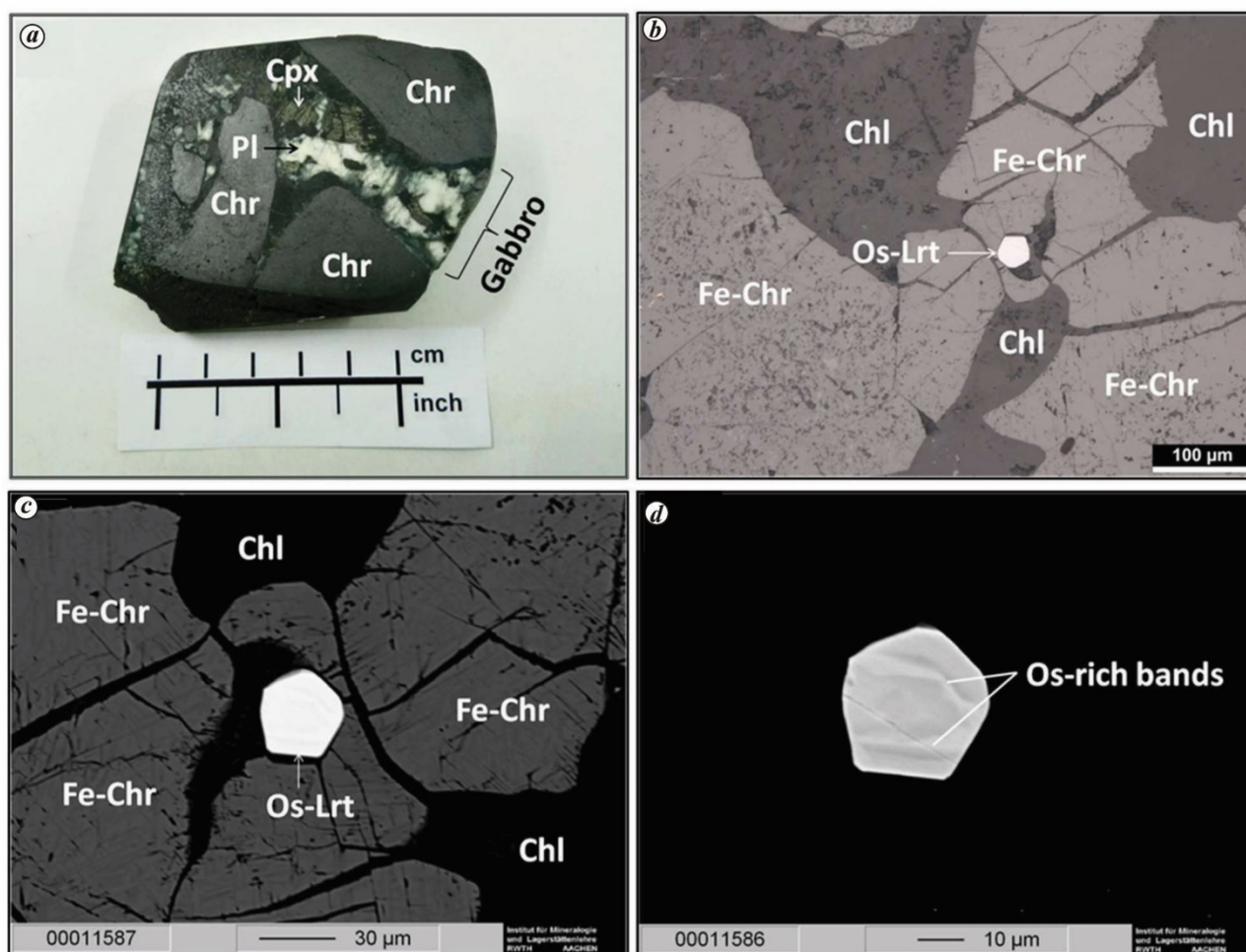


Figure 2. *a*, A brecciated chromite ore sample containing chromite clasts in a gabbro (Bangur gabbro) matrix. Chr, Chromitite; PI, Plagioclase; CPx, Clino-pyroxene. *b*, Reflected light optical microscopic image showing mode of occurrence of osmium-rich laurite (Os-Lrt) in association with ferritchromite (Fe-Chr) and chlorite (Chl). *c*, Back-scattered electron image of a smaller area with an enlarged view of the Os-Lrt grain in electron probe micro-analyzer (EPMA). Note the sharp grain boundaries of Os-Lrt (at least on three sides) shared with ferritchromite. *d*, Enlarged, back-scattered electron image of Os-Lrt (in EPMA) showing the Os-rich bands and the complex oscillatory-type zoning. The other areas have been darkened while capturing the image in order to bring contrast between different bands within the laurite.

(ii) reverse zoning showing Os-rich core (erlichmanite) and Os-poor rim, and (iii) oscillatory zoning having complex intergrowth of Os-rich and Os-poor laurite and/or erlichmanite.

Mondal *et al.*³¹ interpreted a boninitic or high-Mg siliceous basaltic parentage for the chromitites and ultramafic rocks of the Boula-Nuasahi area. This boninitic basalt has compositional similarity with the spatially associated, high-Mg metabasaltic rocks of the Archean greenstone belt belonging to the Iron Ore Group that surrounds the BNUC. Boninitic magmas are formed due to greater amounts of partial melting of metasomatized and severely depleted mantle sources in suprasubduction zones, and are possibly the most sulphur-undersaturated mafic magmas³² having immense potential of forming PGE deposits³³. Generation of boninitic magmas is preceded by the formation and removal of midoceanic ridge basalt (MORB) from the upper mantle. MORB magma generated by a small degree of

partial melting of the mantle is generally sulphide-saturated and shows higher Cu/Pd ratios, leaving the residual mantle with lower Cu/Pd ratios. Therefore, boninitic magmas with lower Cu/Pd ratios are generated by a second-stage melting of the depleted mantle source, which has lost most of its original sulphur/sulphides to the preceding melting event³². Generally, high-Cr chromites [$Cr/(Cr + Al) > 0.70$] that crystallize from boninitic melts in the suprasubduction zone settings are characterized by high $(Os + Ir + Ru)/(Rh + Pt + Pd)$ ratios and have very low Pd + Pt content^{32,34,35}. This PGE pattern is caused by the early extraction of IPGEs (Ru–Os–Ir) with chromite from the magma, leaving behind the more incompatible palladium-group PGEs (PPGEs) (Rh–Pt–Pd) in the melt³⁵. When boninitic magma composed of orthopyroxene, olivine and chromite is formed, the high-temperature PGMs such as laurite or Os–Ir and Pt-alloys crystallize first upon cooling of the melt. This would deplete the IPGE (and also Ni) content in the residual magma and

enrich it with Cu and PPGEs^{36,37}. It has been inferred by Mondal and Zhou¹⁴ that the Bangur gabbro unit of the BNUC has been formed from such Cu and PPGE-enriched fractionated boninitic magma which eventually became sulphur-saturated after emplacement and subsequent metamorphic alterations within the breccia zone.

According to some experimental studies, in an early magmatic stage, Os, Ir and Ru can fractionate and camouflage into chromite, and later on, exsolve from the host chromite to form inclusions of PGM³⁷⁻³⁹. On the other hand, empirical observations⁴⁰ in conjunction with laser-ablation inductively coupled plasma mass spectrometry (LA-ICP-MS) analyses⁴¹ and experimental data⁴²⁻⁴⁴ illustrate that the concentration of Os, Ir and Ru in chromite could be due to physical trapping of submicroscopic clusters of these PGEs in their metallic state, along with larger grains of PGM alloys and sulphides. According to this 'metal clusters' model⁴⁰, PGEs tend to coalesce, forming metallic clusters in the silicate melt. These polyatomic PGE aggregates serve as nuclei for the early-formed chromite and olivine. The segregation of PGE clusters and PGMs from the melt can be attributed to the changing physico-chemical conditions of T, $f(\text{O}_2)$ and $f(\text{S}_2)$ ^{19,45,46}. Accordingly, it is likely that Os, Ir and Ru metallic clusters and alloys could form first, under high T and low $f(\text{S}_2)$ conditions, prior to Ru-rich laurite²². On decreasing T and increasing $f(\text{S}_2)$, the stability field of the alloy and sulphide expands with both alloy and laurite dissolving more Os and Ir. Further decrease in T or increase in $f(\text{S}_2)$ would result in precipitating Os-rich sulphide (erlichmanite) because Os preferentially partitions into sulphide phases upon cooling. However, experimental data^{26,42} indicate

that Os-free laurite and Os-Ir alloys can crystallize together in equilibrium at 1200–1300°C (refs 26, 42) and $\log f(\text{S}_2)$ from –2 to –1.3 in an early magmatic stage. These results, along with thermodynamic data show that the solubility of Os in laurite is more with decreasing T and/or increasing $f(\text{S}_2)$ ^{19,47}. Therefore, the Os-rich laurite crystal (under study) within a chromite grain suggests its crystallization at a slightly lower temperature (<1200°C) and/or higher $f(\text{S}_2)$. Os-Ir alloys were also reported by Augé *et al.*⁸ from the chromitite layers of the peridotite unit from BNUC. These were probably the first to precipitate at high T and very low $f(\text{S}_2)$ in the temperature range 1300–1200°C. Occurrence and origin of smaller laurite crystals in the other Indian ultramafic complexes have been discussed by Meshram and co-workers^{28,29}. Experimental works suggest that laurite can also form during the crystallization of chromite from the basaltic melts at temperatures >1000°C (refs 44, 48). Most Os-poor laurites can crystallize from a sulphur-undersaturated basaltic melt at 1300–1200°C and $\log f(\text{S}_2)$ from –2 to –1.3.

The temperature of stability of pure laurite (RuS_2) is in the range 1300–1150°C (Meshram *et al.*²⁹ and references therein). During progressive crystallization, the temperature gradually decreases with increasing $f(\text{S}_2)$, which facilitates laurite to gradually accommodate more Os and Ir in its lattice during the evolution of the basaltic magma. Meshram *et al.*²⁹ have interpreted that laurite grains displaying patchy zoning in the Chimalpahad anorthosite complex (CAC) chromitites of South India must have formed over a temperatures range (and sulphur fugacity) below the stability of pure laurite, i.e. 1300–1150°C. Since chromite can crystallize below up to a temperature of 1000°C in a late magmatic stage, the possibility of entrapment of any PGM between 1200°C and 1000°C cannot be ruled out. Considering the large size of the laurite grain (~30 µm; which indicates that sufficient time might have lapsed to form this crystal in a 'metals-cluster' model) and the influx of high Os content (~12–15%) into laurite, we interpret that the temperature of crystallization might be <1200°C, otherwise, the laurite could have been very low in its Os content.

Due to the preferential partitioning of Cr^{3+} and Fe^{3+} to chromite, $f(\text{O}_2)$ in the melt around the chromite nuclei may diminish, developing a reduced boundary layer around the chromite nuclei and causing saturation of the most oxidized PGE species (Os, Ru and possibly Ir) in the melt/fluid^{44,49}. Because of their strong affinity to the chromite surface, these PGEs fractionated with chromite will tend to crystallize on the chromite surface, and such physically fractionated PGEs usually occur in the form of extremely fine metallic nuggets of Os, Ru and Ir⁴⁹ and/or submicroscopic rains (or clusters) of Os-Ru alloys which would also incorporate Ir⁵⁰. In such set-ups, these metallic nuggets and submicroscopic alloy clusters stay attached to the boundaries of chromite grains in equilibrium with the surrounding fluid/melt^{42,49}. The coalescence of chromite-bearing melt/fluid bubbles and Os-, Ir- and Ru-bubbles causes a slight

Table 1. Chemical composition of osmium-rich laurite (1 and 2) and its osmium-rich bands (3 and 4) as determined by electron probe micro-analyzer

	1	2	3	4
As	0.214	0.217	0.219	0.200
Pb	nd	nd	nd	nd
S	35.890	35.940	35.550	35.570
Sb	nd	nd	nd	nd
Fe	0.630	0.707	0.626	0.634
Os	12.860	12.910	15.450	15.720
Bi	nd	nd	nd	nd
Te	nd	nd	nd	nd
Co	nd	nd	nd	nd
Ru	45.490	45.672	43.700	43.719
Pd	nd	nd	nd	nd
Ni	nd	nd	nd	nd
Rh	0.806	0.602	0.910	0.876
Cu	0.042	0.023	0.011	0.015
Cd	0.047	0.024	0.000	0.000
Zn	nd	nd	nd	nd
Ir	2.900	2.950	2.520	2.330
Pt	nd	nd	nd	nd
Au	nd	nd	nd	nd
Ag	0.056	0.065	0.088	0.079
Total	98.935	99.110	99.074	99.143

nd, Not detected.

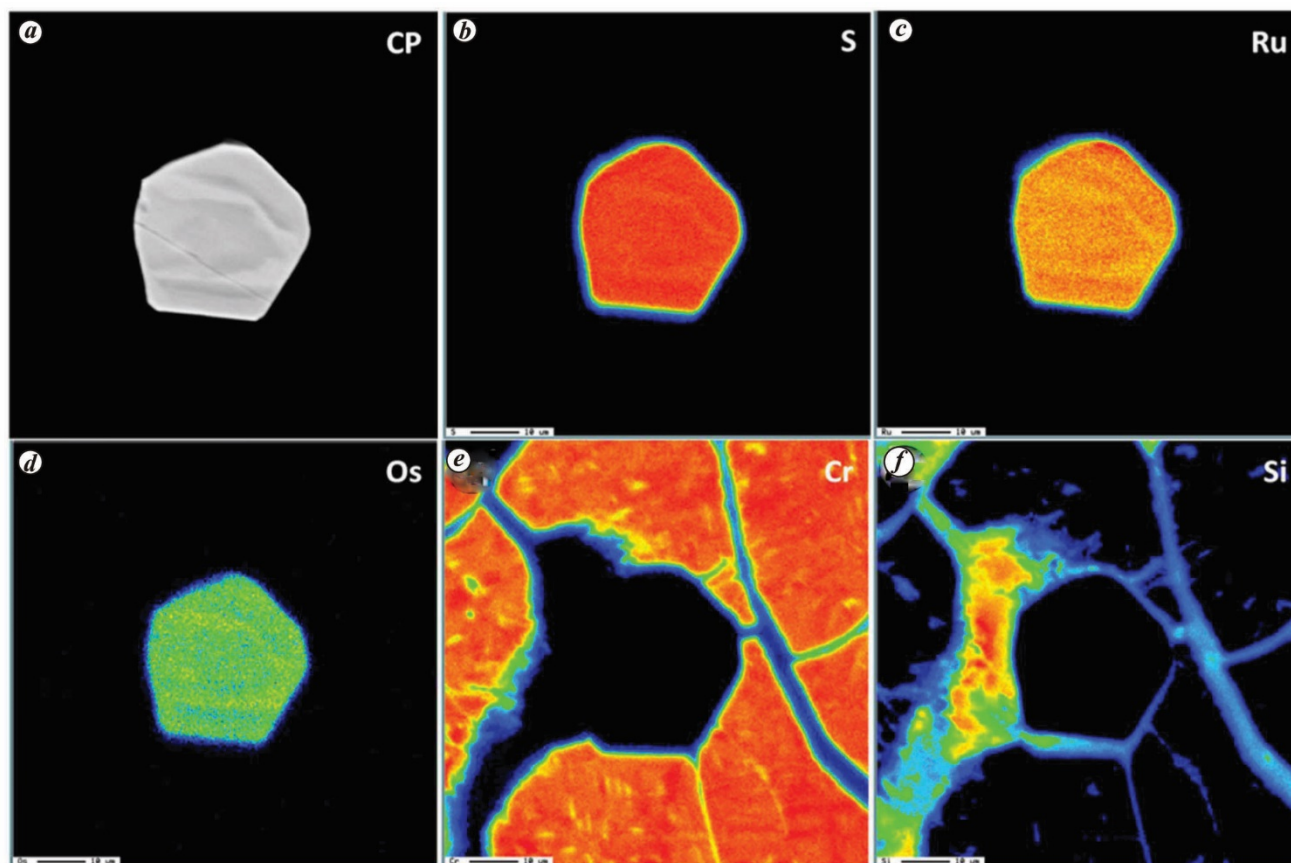


Figure 3. *a*, Back-scattered electron image of laurite in EPMA showing oscillatory zoning. *b–f*, X-ray electron images showing elemental maps for S, Ru, Os, Cr and Si respectively, in the area as in (*a*).

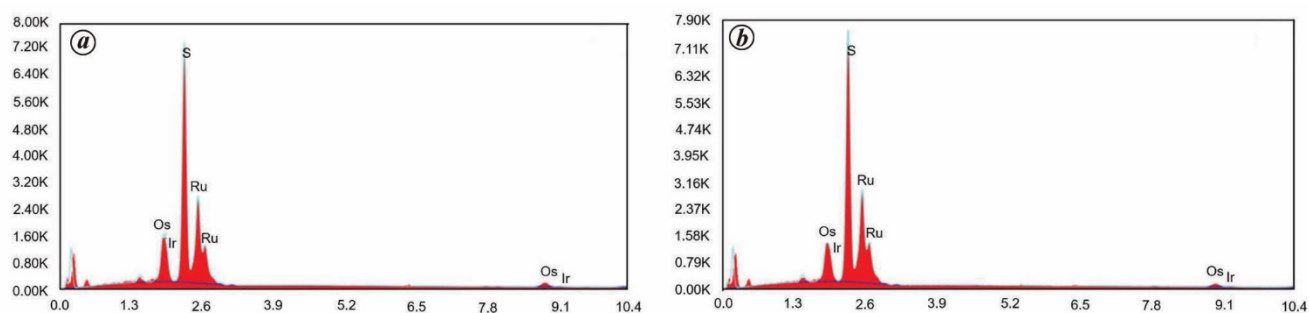


Figure 4. Energy dispersive analytical X-ray peaks of Os-rich bands in (*a*) Os-Lrt and (*b*) that of the Os-laurite main grain showing the peaks of S, Ru, Os and minor signals from Ir. The unnamed peaks in the lower energy level are those of carbon (due to carbon-coating of the sample).

increase in $f(\text{O}_2)$ and $f(\text{S}_2)$, which in turn promotes the crystallization of laurite directly from the fluid/melt or, most likely, by reaction of the previously formed Os–Ir–Ru alloys with dissolved sulphur in the melt⁴². Laurite crystallization by alloy–melt reactions may also take place during hiatuses in chromite growth⁵¹. Thereafter, if the chromite grains start growing again throughout the fluctuations of the turbulent regime, PGMs attached to the edges of chromite get incorporated into the chromite grains⁵¹, prohibiting any further chemical exchange between PGMs and the surround-

ing melt/fluid. The laurite crystallized from alloy–melt reactions is generally Os-poor. When $f(\text{S}_2)$ increases with a decrease in temperature, it gradually accommodates more Os ahead of its entrapment in chromite. After entrapment with the growing chromite, the chromite envelope prevents further exchange of Os from the melt. Consequently, the composition of laurite grains included within the chromite grains reflects the physico-chemical and thermodynamic conditions prevailing during its entrapment, especially the fluctuations in $f(\text{S}_2)$ in the nearby melt. According to this

model, laurite–erlichmanite with different Ru/(Ru + Os) ratios, Os–Ir–Ru alloys, iridium sulphides and irarsite may coexist in different zones of the chromitite body or in a single sample³⁰.

Although it cannot be conclusively mentioned which exact process is responsible for the formation of such a large laurite crystal, from the shape and position of the laurite, it can be reasonably inferred that it was *ab initio* included within the primary chromite grain that had subsequently been hydrothermally altered to ferritchromite. The interstitial chlorite was formed after the dissolution of the associated Fe–Mg silicates that partially replaced the host chromite. Evidences such as euhedral shape, textural association with chromite sharing sharp grain boundaries, and the chemical composition of laurite suggest that laurite had formed during the magmatic crystallization stage. Apart from shape and structure, the composition of this Os–laurite almost matches those found in the type-1 magmatic PGM assemblages with low arsenic content (~0.2 wt%) in contrast to the type-2A and type-2B assemblages, where the laurite is enriched in As (1.01–5.97 wt%, with As substituting for S)⁸. Also, on average, the type-2A and type-2B assemblages contain more Pt and less Ir than laurite in type-1 assemblage. Therefore, this Os-rich laurite appears to have crystallized at a temperature lower than 1200°C in the late magmatic stage.

The osmium content in laurite is considered a direct indicator of the temperature (T)– $f(S_2)$ conditions prevailing during crystallization of the silicate magma. Since $f(S_2)$ increases with decreasing temperature (upon cooling) in a magmatic system^{19,52–54}, normal patterns of zoning, i.e. Os enrichment towards the rim, in laurite–erlichmanite grains would record different steps of their crystallization history prior to their entrapment by chromite. However, this continuous decrease in temperature and increase in $f(S_2)$ cannot explain the reverse and oscillatory zoning patterns found in some laurite–erlichmanite grains^{22,52}. The Os–laurite grain in our sample showed complex oscillatory (patchy) zoning with some Os-enriched bands (Figures 2 d and 3). The brighter bands were enriched in Os (~15 wt%) compared to other areas in the grain (Os ~12 wt%). Such chemical variations need to be explained by a mechanism that can generate different $f(S_2)$ gradients in space and/or time, which may or may not include temperature changes in a short period of time before entrapment of the PGMs by chromite. Variable $f(S_2)$ gradients could be possible during the crystallization of chromite by magma mixing^{50,55–57} or as a consequence of the segregation of a fluid phase from the parent melt⁵⁸. According to Mondal *et al.*³¹, a combination of magma mixing involves a boninitic or high-Mg siliceous basaltic magma, with a relatively high water content of the mixed magma in a supra-subduction zone setting are the possible factors for the origin of chromitites in the Nuasahi complex. According to the model proposed by González-Jiménez *et al.*³⁰, such zoned crystals are generated under changing thermodynamic conditions. In such a case, chromite crystallization occurs in a dynamic setting charac-

terized by mixing successive batches of olivine–spinel saturated melt in a chamber hosting a more differentiated melt^{50,55,57}. The sequential melt injections and the temperature contrast between the peridotite wall-rock and the melt, cause the magmas to mingle in a turbulent regime. Sharp changes in physico-chemical conditions are also possible during the crystallization of chromite by magma mingling with or without segregation of a fluid-rich fraction. The turbulent regime promotes the coalescence of chromite-rich melt bubbles, which leads to joint chromite forming larger crystals. Such a system can be generated by a periodic supply of melt batches within a conduit, like that proposed by Lago *et al.*⁵⁹. The denser chromite-bearing bubbles are transported to the walls of the conduits⁵⁸, like in the elutriation cell described by Lago *et al.*⁵⁹, promoting their coalescence and partial solidification. Each melt bubble will retain its own $f(S_2)$, $f(O_2)$ and chemical composition till it coalesces with a different bubble. This could produce the variable heterogeneous environment essential to create the different patterns of zoning in laurite–erlichmanite grains.

Post-magmatic alteration, such as serpentinization, can also produce zoning in laurite–erlichmanite^{60–62}. Inversion of magmatic sulphur fugacity trend can occur during weathering at low temperatures and serpentinization, i.e. $f(S_2)$ decreases favouring the formation of native PGEs in most cases rather than sulphides. Especially the magmatic laurite–erlichmanite can be altered *in situ* by progressive sulphur loss and adding base metals and possibly oxygen, generating secondary PGE alloys or oxides^{19,63}. During the post-magmatic stage, when $f(S_2)$ decreases, the primary laurite can adjust its composition by releasing Os and probably Ir, which form secondary alloys⁶⁴, resulting in zoning⁶⁵. The laurite grains affected by serpentinization are usually anhedral to subhedral with the overgrowth of Ru oxide or hydroxide⁶⁴ and/or Os–Ir alloys⁶⁵ on the primary laurite. However, in the present case, the euhedral shape shared boundaries with the chromite grain and the absence of any such overgrowth of Ru oxide or hydroxide and/or Os–Ir alloys are not in accordance with the zoning formed due to serpentinization.

The ferritchromite and chlorite (associated with laurite) are supposed to have been formed from pristine magmatic chromite and Fe–Mg silicates respectively, by post-magmatic hydrothermal alteration (stage-2 to stage-4, ~500–600°C, as mentioned by Augé and Lerouge⁹), related to the Bangur gabbro intrusion. However, the euhedral shape, typical magmatic composition, and no overgrowths indicate the magmatic origin of the laurite grain. This study also suggests that the complex oscillatory (patchy) zoning in the laurite grain had formed in the magmatic crystallization stage due to local variation of thermodynamic conditions. Although the host ferritchromite grain is a product of the post-magmatic (high temperature) hydrothermal alteration (500–600°C), the laurite grain has not been affected by this alteration process.

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