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LETTER

PGE distribution in massive sulfide deposits of the Iberian Pyrite Belt

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Abstract We present the first platinum group elements (PGE) data on seven massive sulfide deposits in the Iberian Pyrite Belt (IPB), one of the world largest massive sulfide provinces. Some of these deposits can contain significant PGE values. The highest PGE values were identified in the Cu-rich stockwork ores of the Aguas Teñidas Este (Σ PGE 350 ppb) and the Neves Corvo (Σ PGE 203 ppb) deposits. Chondrite normalized PGE patterns and Pd/Pt and Pd/Ir ratios in the IPB massive, and stockwork ores are consistent with the leaching of the PGE from the underlying rock sequence.

Keywords Platinum group elements · Massive sulfide deposits · Iberian Pyrite Belt · Spain · Portugal

Introduction

Layered igneous complexes are the principal source of platinum group elements (PGE). However, significant PGE enrichment has also been reported from ancient volcanic-hosted massive sulfide deposits (e.g., Pan and Xie 2001; Vikent'ev et al. 2002) and their modern analogues (Crocket 1989, 1990; Cave et al. 2003; Pašava et al. 2004). We are unaware of any reports on PGE in massive sulfide deposits of the Iberian Pyrite Belt (IPB) that belongs to one of the largest sulfide provinces in the world.

The main purpose of this contribution is to report the results of a study on the distribution of PGE in seven massive sulfide deposits of the IPB (Aljustrel, Aguas Teñidas, Rio Tinto, San Miguel, San Platón, Tharsis, and Neves Corvo), to compare them with similar data from recent and ancient analogues, and to discuss them towards a better understanding of the metal sources and genetic processes involved in the formation of massive sulfide deposits in this remarkable province.

Geological setting

The Variscan Belt of South Western Iberia includes two very different terranes, the South Portuguese Zone and the Ossa Morena Zone. These terranes were accreted during the Variscan orogeny and have contrasting geologic and metallogenic evolutions (Quesada 1991; Tornos et al. 2005; Fig. 1). The South Portuguese Zone includes the IPB that consists of a late Devonian–Carboniferous sequence dominated by siliciclastic rocks (phyllite–quartzite group, base unknown) overlain by a relatively thin (generally less than 600 m) volcano–sedimentary sequence (volcanic–sedimentary complex) and a thick synorogenic turbiditic sequence. The IPB is one of the largest massive sulfide provinces in the world hosting more than 85 known deposits, out of which eight have total tonnages in excess of 100 million metric tonnes (Mt). The total resource is more than 1,700 Mt of massive and stockwork ores (Leistel et al. 1998; Tornos 2006). The deposits are hosted by the late Devonian to the early Carboniferous volcanic–sedimentary complex, which consists of a heterogeneous submarine succession dominated by rhyolitic to dacitic volcanic rocks

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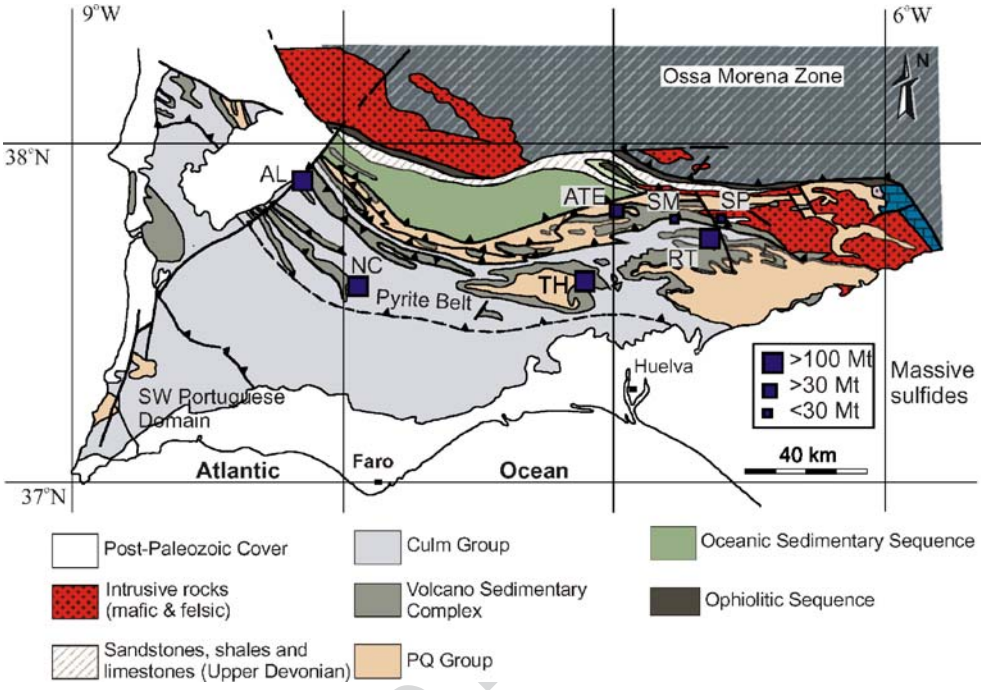


Fig. 1 Geological setting of SW Iberia showing the location of the sampled ore deposits. *AL* Aljustrel, *ATE* Aguas Teñidas Este, *NC* Neves Corvo, *RT* Rio Tinto, *SM* San Miguel, *SP* San Platón, *TH* Tharsis. Modified from Tornos (2006)

t1.1 **Table 1** Description of studied samples from the Iberian Pyrite Belt

t1.2	Deposit	Sample	Sample description
t1.3	Aguas Teñidas zn–pb–cu (ag) massive sulfide and stockwork deposit hosted by massive to fragmental rhyolite and	$AT7_{MS}$	Cu-rich massive sulfides from hanging wall, stope 947
		$AT8_{MS}$	Zn–Pb-rich massive sulfides-banded sphalerite and galena
t1.4	rhyo–dacite; highly deformed		with minor pyrite and chalcopyrite, stope 925
t1.5	San Miguel replacive pyrite-rich massive sulfides and related stockwork in dacite	$AT9_{MS}$	Massive sulfides-dominated by Au-rich pyrite, stope 952
		$SM10_{MS}$	Massive sulfides-dominated by pyrite with fragments of host
t1.6			rock
t1.7	San Platón Zn–Pb–Cu (Ag–Au) massive sulfide deposit,	$SP11a_{MS}$	Massive sulfides-banded sphalerite
t1.8	replacive on pumice breccia	$SP11b_{MS}$	Massive sulfides-dominated by pyrite
t1.9	Tharsis exhalative shale-hosted pyrite-rich massive sulfide deposit with a stockwork enriched in Au–Co–Te–Bi	$Th12_{MS}$	Pyritic ore. Massive sulfides of the Filón Norte orebody
		$Th13_{MS}$	Carbonate ore. Pyrite–siderite-rich massive sulfides from the
t1.10			San Guillermo orebody
t1.11		$Th14_{ST}$	Pyrite-rich stockwork underlying the San Guillermo orebody
	Neves Corvo High Cu and Sn-grade, replacive massive sulfide	$NC15MC_{MS}$	Massive Cu-rich sulfides from the immediate footwall of the
t1.12	deposit hosted by shales and rhyolitic to dacitic volcanic and		massive orebody, remobilized chalcopyrite ore
t1.13	volcaniclastic rocks with underlying Cu-rich and massive cassiterite-rich stockworks (Relvas et al. 2006)	$NC16MC_{MS}$	Massive Cu-rich sulfides from the massive orebody
		$NC17Mpy_{MS}$	Massive pyrite from the upper part of Cu-rich massive
t1.14			orebody
		$NC18Zn_{MS}$	Massive Zn-rich sulfide from the footwall of the massive
t1.15			Cu-rich orebody
		$NC19RC_{ST}$	"Rubané" Cu-rich stockwork mineralization tectonically
t1.16			deformed and emplaced on top of the Corvo orebody
t1.17		NC20jasp	Jasper sample (marker of hanging wall)
t1.18		$NC21Sn_{MS}$	Massive Sn-rich sample, C 778 1T
t1.19	Aljustrel Zn–Cu–Pb-rich massive sulfide deposit in fragmental	$Alj22Mpy_{ST}$	Pyrite from the stockwork underlying the massive sulfides
t1.20	felsic volcanic rocks	$Alj23Mpy_{MS}$	Pyrite-rich massive sulfides
	Rio Tinto Pyrite-rich massive sulfide deposit hosted by shale	$RT24_{MS}$	Replacive pyrite-rich massive sulfides hosted in dacite from
t1.21	with underlying pyrite and Cu-rich stockwork and replacive massive sulfides hosted by massive and fragmental dacite		the Filon Norte orebody
		$RT25_{ST}$	Pyrite-rich stockwork hosted by dacite of the Filon Norte
t1.22			orebody

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and subordinate basalts that occur interbedded with abundant mudstones and chemical sediments (Schermerhorn 1971; Oliveira 1990; Leistel et al. 1998). Shale-hosted massive sulfide deposits dominate in the southern part of the belt and are characterized by, on average, larger tonnages, lower base metal contents, a significant input of biogenic sulfur, and the common presence of synsedimentary features; they have been interpreted as formed in small anoxic basins. The deposits in the northern part of the belt are mostly hosted by porous/reactive glass- and pumice-bearing felsic volcanic rocks, show a systematic metal zoning, and have been interpreted as formed by subseafloor replacement of volcanic rocks (Tornos 2006).

Methods

A total of 20 samples of 1 kg each of representative ore types from seven massive sulfide deposits were studied for their PGE and Au contents (Table 1). The location of the sampled deposits is shown in Fig. 1. A brief description of the main characteristics of the samples and a short note on the geological setting of each deposit have been included in Table 1. The samples were pulverized in the laboratories of the Czech Geological Survey (Prague). The PGE and Au were preconcentrated into Ni-sulfide and determined by inductively coupled plasma atomic emission spectrometry in the ALS Chemex laboratories (Brisbane, Queensland, Australia). Detection limits are as follows: Pt, Pd, Ir, Os, Rh, Ru, 2 ppb; and Au, 5 ppb. All Ir and Os values are below the detection limit of 2 ppb (Table 2).

Results

The results of PGE determinations are presented in Table 2 and shown in Fig. 2a–c. The samples define the means and the 1-sigma standard deviations of 49±47 ppb Pt and 32± 26 ppb Pd, with Pd/Pt=0.77 (range, 0.37–1.5). The highest Pt (201 ppb) and Pd (116 ppb) values were detected in a Cu-rich stockwork sample (Cu=3 wt%) of the Aguas Teñidas Este deposit that also shows the highest sum of PGE (350 ppb). The PGE contents in this deposit decrease from the Cu-rich ore through the Zn–Pb-rich polymetallic ore to the Au-rich pyrite sample (Au=955 ppb). The maximum concentration of Ru (26 ppb) was detected in a massive sulfide pyrite-rich sample from the San Miguel deposit, and the highest Rh content (27 ppb) was found in a pyrite-rich sample from stockwork at the Tharsis deposit. At the Neves Corvo deposit, the highest Pt (126 ppb) and Ru (18 ppb) contents were detected in the "Rubané" Cu-rich ore, whereas the maximum Pd (60 ppb) and Rh (13 ppb) values were found in the massive pyrite sampled above the Cu-rich base of the massive sulfide orebody. The Pd/Au ratios of the massive sulfide deposits vary from 0.018 to 0.563.

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t2.1 **Table 2** PGE and Au data for studied samples, Iberian Pyrite Belt

t2.2	Deposit	Sample	Pt (ppb)	Pd (ppb)	Rh (ppb)	Ru (ppb)	Au (ppb)	Pd/Pt	Pd/Au
t2.3	Aguas Teñidas	AT7	201	116	13	21	498	0.58	0.233
t2.4		AT8	84	37	8	12	378	0.44	0.098
t2.5		AT9	24	17	<2	9	955	0.71	0.018
t2.6	San Miguel	SM10	52	34	12	26	64	0.65	0.531
t2.7	San Platon	SP11a	8	12	4	3	>1,000[a]	1.50	<0.012
t2.8		SP11b	36	18	<2	8	>1,000[a]	0.50	<0.018
t2.9	Tharsis	Th12	13	9	<2	8	>1,000[a]	0.69	<0.009
t2.10		Th13	36	22	<2	9	>1,000 [a]	0.61	<0.022
t2.11		Th14	40	20	27	12	191	0.50	0.105
t2.12	Neves Corvo	NC15MC	21	22	9	9	177	1.05	0.124
t2.13		NC16MC	91	58	9	16	103	0.64	0.563
t2.14		NC17Mpy	84	60	13	9	198	0.71	0.303
t2.15		NC18Zn	39	30	8	6	367	0.77	0.082
t2.16		NC19RC	126	47	12	18	285	0.37	0.165
t2.17		NC20jasp	23	15	8	7	126	0.65	0.119
t2.18		NC21Sn	10	11	<2	6	148	1.10	0.074
t2.19	Aljustrel	Alj22Mpy	57	69	9	13	>1,000[a]	1.21	<0.069
t2.20		Alj23Mpy	13	12	<2	8	173	0.92	0.069
t2.21	Rio Tinto	RT24	20	14	<2	6	>1,000[a]	0.70	<0.014
t2.22		RT25	7	8	9	6	>1,000 [a]	1.14	<0.008
t2.23	Average		49	32	7	11	533	0.77	0.132

t2.24 [a] Au values are in excess of ICPAES maximum detection limit (1 000 ppb).

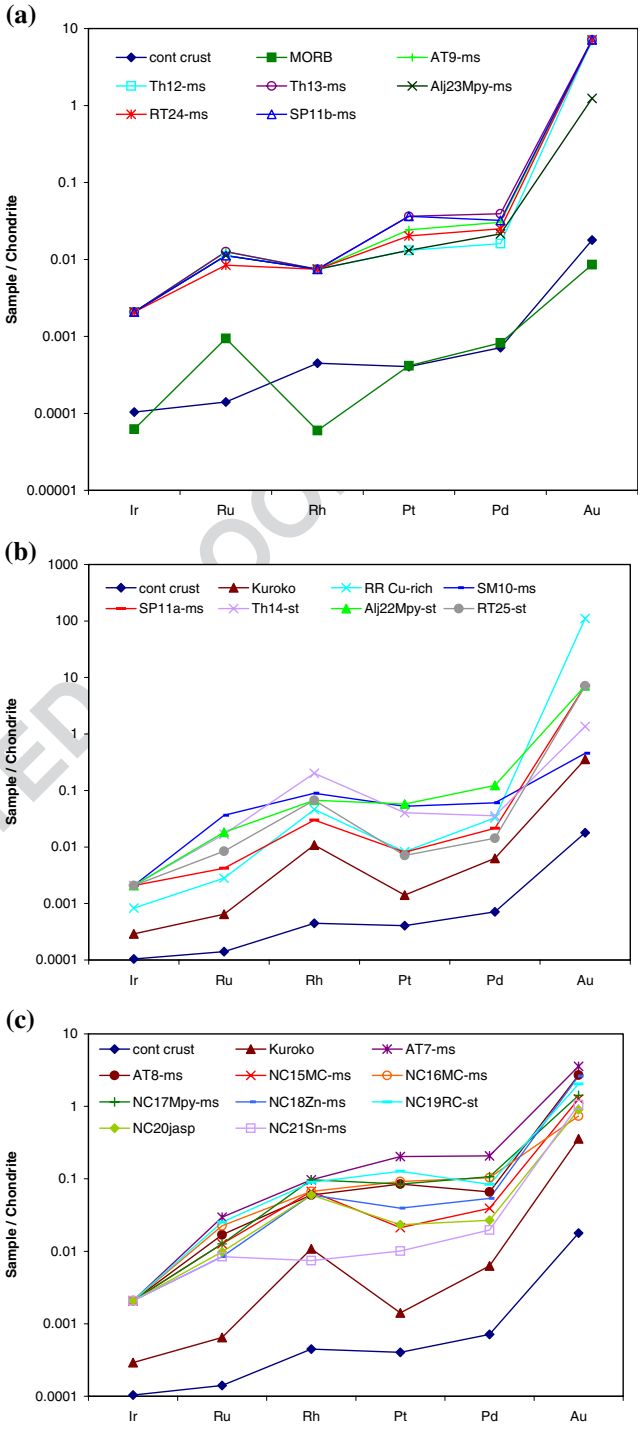


Fig. 2 Chondrite-normalized PGE–Au patterns **a** for pyrite-dominated shale-hosted massive sulfide ore from Tharsis (Th12, Th13), pyrite-dominated volcanic-hosted massive sulfide ore from Aljustrel (Alj23 Mpy), Aguas Teñidas (AT9), San Platón (SP11b), and Rio Tinto open pit (RT24) compared to the Cu-rich sample from the Roman ruins (RR Cu-rich sample), MORB, and global continental crust; **b** for pyrite-rich stockworks hosted by dacitic rocks from Aljustrel (Alj22Mpy), Rio Tinto–Quebrantabuesos (RT25), Tharsis–San Guillermo orebody (Th14), Zn-rich sample from San Platón (SP11a), and massive sulfide sample from San Miguel (SM10) compared to the Kuroko deposits and continental crust; **c** for samples from Neves Corvo and Aguas Teñidas (AT7 and AT8) compared to the continental crust and Kuroko deposits. The MORB data are from Crocket (2002), the continental crust data are from the GERM reservoir database (http://www.earthref.org/), the Kuroko data from Pan and Xie (2001), the Roman ruins (Pacmanus) data from Pašava et al. (2004), and the chondrite data from Anders and Grevesse (1989). *ms* Massive sulfide, *st* stockwork

Discussion

Volcanic-hosted massive sulfide deposits often have significant PGE contents, although the current database is very limited. For example, values as high as 1,789 ppb Pd and 817 ppb Rh have been reported in the pyrite–chalcopyrite-rich banded ore from Besshi (Shikoku, Japan; Pan and Xie 2001).

The most pronounced feature of the PGE distribution in the massive sulfide deposits of the IPB is the enrichment in Pt, Pd, and Rh in the copper-rich ore of both Aguas Teñidas and Neves Corvo; this enrichment is significantly higher than that observed in the associated polymetallic, tin-rich, and pyrite gold-bearing samples (Table 2). Similar values to those reported in this paper (Pt and Pd in the range of 10 to 20 ppb, with Pd/Pt<1) have also been found in volcanic-hosted massive sulfide deposits of the Urals (Vikent'ev et al. 2002). These results are partially consistent with those of Pašava et al. (2004) who documented a significant positive correlation between Cu and Pd and Rh (but not with Pt) in the massive sulfide mineralization of the active hydrothermal field at the Pacmanus basin.

Two different types of chondrite-normalized PGE patterns can be distinguished in our samples. The first group of samples shows a slightly negative Rh anomaly similar to the mid-ocean ridge basalt (MORB) pattern (Fig. 2a); it includes data from most massive sulfide ore samples coming both from the shale-hosted deposits (Tharsis and Rio Tinto) and from several volcanic-hosted ones, such as Aljustrel, Aguas Teñidas, or San Platón. The second group of samples (Fig. 2b) is characterized by lower PGE contents, a positive Rh anomaly, and includes the stockworks of Aljustrel, Rio Tinto, and Tharsis, as well as a sample of the Zn-rich ore of San Platón. The PGE fractionation in these samples is comparable to that of continental crust and very similar to the average pattern for Kuroko-type deposits (Pan and Xie 2001) and for Cu-rich samples from Roman ruins, an active hydrothermal field in the Pacmanus basin (Pašava et al. 2004). In this modern analogue, the PGE were interpreted to have been leached from mixed sources (dominantly felsic volcanic and sedimentary rocks). Consistently, the Pd/Au values reported for the Kuroko ores (0.004 to 0.07; Pan and Xie 2001) and for the average continental crust (0.16, data from the geochemical earth reference model reservoir database, http://www.earthref.org) fall within the range of values yielded by this subset of samples (0.105–0.531).

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Some Cu- and Zn-rich samples (NC-17Mpy, NC-18Zn, NC-19RC, and NC-20jasp-see Table 1) from the Neves Corvo deposit display a slightly positive Rh anomaly similar to that of samples from group 2, whereas the remaining two (Cu-rich) samples from this deposit (NC-15MC and NC-16MC) show a rather flat chondrite-normalized PGE pattern—apparently a transition between the patterns displayed by the two sample groups (Fig. 2c). The samples AT-7 (Cu-rich) and AT-8 (Zn–Pb rich) from the Aguas Teñidas deposit show a similar shape of chondrite-normalized PGE patterns. The PGE distribution of the tin-rich ore from Neves Corvo (sample NC21Sn, Fig. 2c) is very similar to the first group of pyrite-rich massive sulfides samples (Th12, Alj23Mpy, and RT24; Fig. 2a).

Possible sources of PGE in the ore-forming fluids of the IPB include the underlying siliciclastic sequence (phyllite–quartzite group), the felsic volcanic rocks, or their unexposed plutonic roots (Relvas et al. 2001; Tornos et al. 2005). In the absence of the PGE data from the felsic rocks from the IPB, an average value of 4 ppb Pd, 6 ppb Pt, and 0.2 ppb Ir ($Pd/Pt_{avg.}=0.7$) was assumed on the basis of the published data for rift-related felsic volcanic rocks (closest type of rocks with available PGE analyses; Borg et al. 1988; Barnes et al. 1988). Our results show that the Pd/Pt ratios of the analyzed IPB ores are broadly similar to the Pd/Pt ratio estimated for these rocks (Table 2). It is difficult to estimate whether the Pd/Pt ratio reflects the situation in the original source rock or whether the interaction with the country rocks occurred during the ascent of hydrothermal fluids and/or at the site of sulfide precipitation, or even resulted from the mixing of fluids with different sources. However, the fact that most of the massive sulfide ore samples have patterns that are different from the ones yielded by the underlying stockwork ores suggests that the PGE contents should have been controlled both by the physics and chemistry of the ore-forming fluid and by factors related to the conditions of precipitation (e.g., fluid mixing with unmodified/less modified seawater, biogenic activity, redox regime). A similar two-source model has been proposed for the sulfur in these massive sulfide deposits (Velasco et al. 1998; Tornos et al. 2005). The PGE contents of the stockwork samples are more likely to be inherited from the host rock, suggesting that the ore-forming fluids were equilibrated with the felsic igneous rocks and/or the metasediments from the phyllite–quarzite group and/or their unknown underlying basement. Radiogenic isotope geochemistry is consistent with such a model because the Os, Pb, and Sr isotope data indicate that the fluids were equilibrated with the old crustal continental rocks (Marcoux 1998; Munha et al. 2005; Tornos et al. 2005). Largely, a crustal source of metals for Tharsis and Rio Tinto pyrite-rich samples was suggested by Mathur et al. (1999) based on the Re–Os isotopes. The fluid inclusion data suggest that fluids involved in the massive sulfide ore-forming processes were aqueous with variable but usually high salinities, ranging on average between 3- and 12-wt% NaCl eq. and temperatures up to 300°C (Aguas Teñidas Este, 82–240°C; San Miguel, 160–240°C; Neves Corvo, 112–196°C; Rio Tinto, 140–260°C, Tornos 2006; Tharsis ore, below 250°C, Kase et al. 1990). Such fluids are able to transport the PGE as chloride complexes, as documented by Gammons et al. (1992), Wood et al. (1992), and others.

Conclusions

The study of the distribution of PGE in the volcanic-hosted massive sulfide deposits of the IPB (South Portuguese Zone) can be summarized as follows:

1. The IPB ores analyzed are characterized by average Pt (49 ppb), Pd (32 ppb), and Pd/Pt (0.77). The highest Pt and Pd values were identified in the Cu-rich stockwork ore of the Aguas Teñidas and Neves Corvo deposits. Pyrite-, Zn-, Pb-, and Sn-rich samples generally show lower PGE values.
2. The PGE patterns of the massive sulfide ores are different from those of the underlying stockwork ores, suggesting that the fractionation of the PGE may have been conditioned not only by the physics and chemistry of the ore-forming fluids, but also by factors related to the environment of deposition, such as mixing with unmodified seawater, biogenic activity, or others.
3. The chondrite-normalized PGE patterns and Pd/Pt ratios of the massive sulfide and stockwork ore samples studied suggest that most PGE contained in the IPB ores should have been leached from the felsic volcanic and metasedimentary rocks hosting the deposits, their plutonic equivalents, and/or the siliciclastic rocks of the underlying phyllite–quartzite group.

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