Mineralogical Magazine, August 1999, Vol. 63(4), pp. 595–602

Letter

Magmatic gold grains in the El Tale lamproite, Fortuna, SE Spain

L. Toscani

Dipartimento di Scienze della Terra, Università di Parma, Parco Area delle Scienze 157A, 43100 Parma, Italy

ABSTRACT

Gold of magmatic origin has been discovered in the lamproitic dyke of El Tale (Fortuna, Province of Murcia), a small outcrop of ultrapotassic rocks of the Tertiary magmatic province of southeastern Spain. It is the first finding of gold grains in lamproite magmas from southeastern Spain, and indeed worldwide. Pure gold occurs only in the glassy groundmass as very rare spherules and globular grains (up to 5 μm). Despite the occurrence of gold grains, the bulk rock contains not more than ~19 ppb Au.

The magmatic origin of gold is inferred from the heterogeneous distribution of the grains in the rock, the shape of the grains (which is comparable to that of gold grains crystallized experimentally from mafic liquids) and from the As-Sb-S-Cl-poor composition of the bulk rock. The Pd/Ir and Au/Pd ratios, little affected by the early segregation of olivine (~10 vol.%), support an Au- and PGE-rich composition of the mantle source. The low f_{O_2} ($\approx$IW) and the complexing with F and alkalies probably favoured partitioning of Au and PGE into the lamproite magma.

Keywords: gold, platinum-group elements, lamproite, SE Spain.

Introduction

In the last 20 years, ultrapotassic lamproites have been investigated widely for their unusual mineralogical and geochemical characters. The discovery of diamonds in the lamproites of Argyle (East Kimberley) and Ellendale (West Kimberley), Western Australia, increased the economic interest in these rocks. Noble metals have not generally been investigated in the lamproitic rocks. However, data for Au and PGE are reported for the diamondiferous lamproites of Western Australia (Lewis, 1987; Jaques, 1990), the Archaean lamprophyres of the Yilgarn Block, also in Australia (Taylor *et al.*, 1994) and, for some Canadian lamprophyres (Superior Province, Kerrich and Wyman, 1994).

In this paper we describe microscopic gold grains of probable magmatic origin occurring in the ultrapotassic lamproite of El Tale (Fortuna, southeastern Spain) and report the first data on the Au and PGE content of Spanish lamproites.

The dyke of El Tale

The lamproite of Fortuna (province of Murcia) forms three small outcrops: the neck of Cabecitos Negros (typical 'fortunite'; Fuster *et al.*, 1967); the dyke of Los Derramadores; and the dyke of El Tale (Fig. 1). The dyke of El Tale is 600 m long and 10 m thick and oriented approximately E–W and crops out ~3.5 km east of the village of Fortuna. It cuts the Tertiary sediments (marly limestones, marls and clays) subvertically. The rock is fresh and consists of phlogopite (Mg# = 90) phenocrysts, minor olivine (Mg# = 91 and containing Cr-spinel), clinopyroxene and apatite in a vitreous matrix. In the central part of the dyke, the matrix may contain laths of sanidine. The rocks underwent ~10% olivine fractionation (Toscani and Salvioli-Mariani, submitted). Postmagmatic alteration, with deposition of Ni-rich sulphides, affected only the easternmost part of the dyke.

The dyke has been sampled in all parts. The rock, which exhibits a rather constant bulk

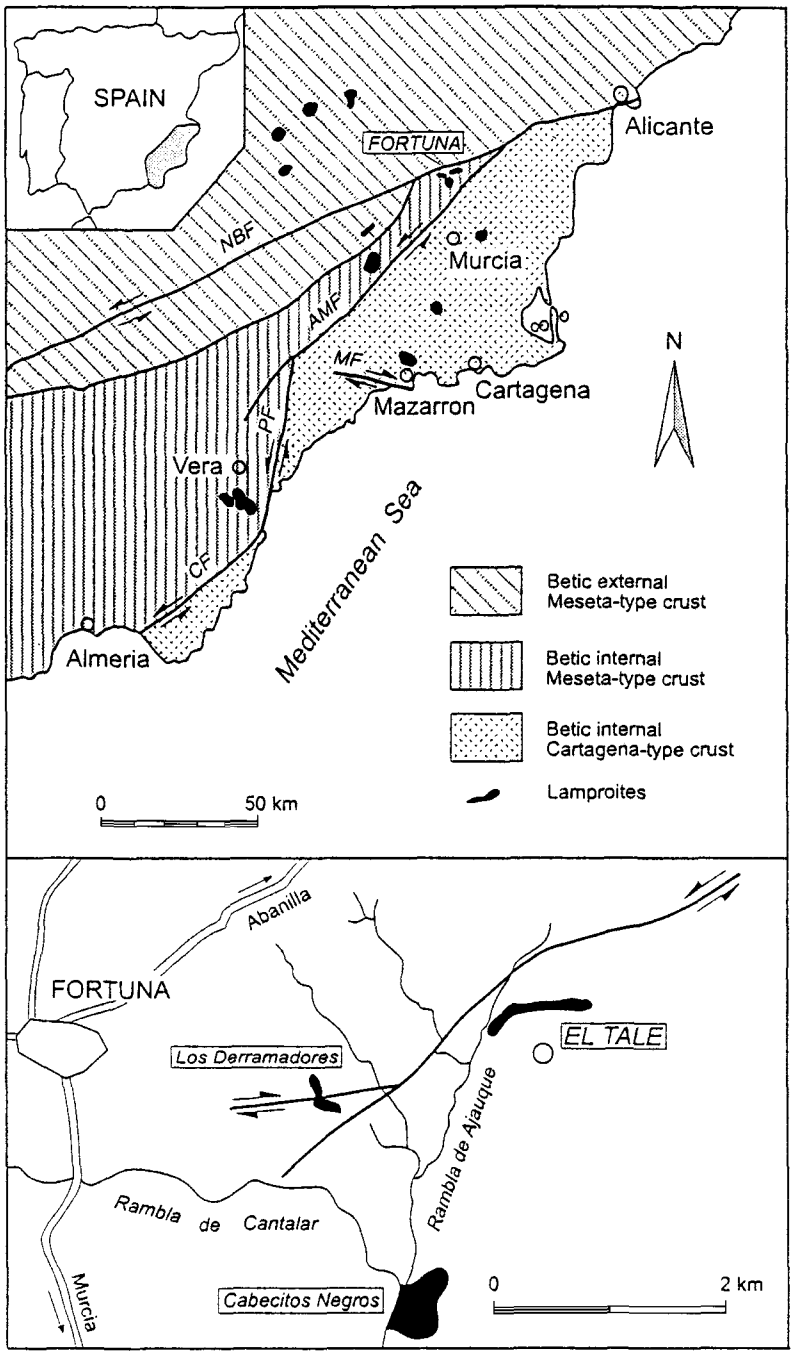

FIG. 1. Geological sketch map of southeastern Spain and the lamproite outcrops of Fortuna (after Fuster *et al.*, 1967, partially modified). The denomination of the types of continental crust is after De Larouzière *et al.* (1988). Main Neogene faults: NBF = North Betic Fault; AMF = Alhama de Murcia Fault; PF = Palomares Fault; MF = Mazarron Fault; and CF = Carboneras Fault.

composition, is silica-oversaturated, metaluminous (Agpaitic Index A.I. = (Na + K)/Al = 0.93–0.95, atomic ratio), ultrapotassic (Foley *et al.*, 1987) with high LILE, HFSE and *REE*, relatively low MgO and Ni, and low Na_2O, CaO and Sc (Table 1). The oxygen fugacity of the magma, calculated according to the method reported in Venturelli *et al.* (1988), was low (IW+1.0 to 1.6 log unit). Fluorine (F $\approx$ 0.5 wt.%; Table 2) reaches the highest values found in Spanish lamproites (Contini *et al.*, 1993) and in lamproite rocks worldwide (cf. Sekerin *et al.*, 1993; Jaques *et al.*, 1986), whereas Cl is low (0.09 wt.%). Carbon dioxide is fairly constant and low ($\approx$0.25 wt.%), with the exception of sample SP839, where secondary carbonate is present. Sulfur is typically low ($\leqslant$0.03 wt.%), but in the sulphide-bearing portion of the dyke reaches 0.12 wt.%. Both As and Sb are low (Table 1), and do not correlate with Au. Sample SP839 exhibits relatively high As (48 ppm) and Sb (690 ppb) contents. The As and Sb are trace elements usually found in association with gold in mesothermal gold deposits; this, however, is not the case with sample SP839 where the Au content is comparable to that of other samples of the dyke.

Gold

Gold occurs as spherules and multi-globular grains only in the glassy, unaltered groundmass. The grains are very rare and distributed heterogeneously at the millimetre scale. The shape and size of the grains correlate: the smallest grains (tenths of μm, Fig. 2 *e,f*) are invariably spheroidal whereas the multi-globular grains reach the largest size (up to 5 μm; Fig. 2*b*). This correlation suggests a prompt nucleation from the liquid and a more extended growth for the smallest and the largest grains respectively.

Semi-quantitative (SEM-EDS) analysis indicates that the grains are pure gold in composition. The peaks of Au are sharp and symmetrical, suggesting that the adjacent energy lines of Pt are absent or negligible. Silver is below the detection limit (Ag <0.5 wt.%).

The finding of gold has been fortuitous and the gold grains have been detected in three samples (SP835, 839, 842) from the mid-, and mid–east sections of the dyke. However, the peculiar distribution of the grains in the rock and the investigation of only a few thin sections per sample means we cannot rule out the possible occurrence of gold grains in the other parts of the dyke.

TABLE 1. Representative bulk rock composition of the ultrapotassic El Tale lamproite (sample SP842): after Toscani and Salvioli-Mariani (submitted)

SiO_2	59.14	V	110	Pb	62	La	72.6
TiO_2	1.46	Sc	16	Th	112	Ce	222
Al_2O_3	12.42	Cr	629	U	15.9	Nd	141
Fe_2O_3	4.82	Ni	163	Ga	24	Sm	25.6
MnO	0.04	Co	26	Ta	2.83	Eu	3.94
MgO	5.96	Cu	37	Hf	20.6	Gd	14
CaO	2.71	Zn	82	Ba	1587	Tb	1.88
Na_2O	1.30	Rb	828	Cs	11.4	Tm	0.41
K_2O	8.73	Sr	493	As	23	Yb	2.14
P_2O_5	1.2	Y	19	Sb	395	Lu	0.26
LOI	1.7	Nb	37				
Total	99.4	Zr	753				
A.I.	0.93						
Mg#	71						

Oxides as wt.%
Elements as ppm except Sb (ppb)
Agpaitic Index A.I.= (Na + K) / Al (atomic)
Mg# = 100 * Mg / (Mg + Fe) (atomic)

TABLE 2. Bulk rock analyses of the ultrapotassic El Tale lamproite

	SP835	SP838	SP839	SP842	SP844	SP845
CO_2	0.32	0.22	3.35	0.34	0.25	0.13
F	0.47	0.46	0.47	0.48	0.5	0.48
Cl	855	1735	895	665	920	815
S	0.017	0.025	0.022	0.014	0.055	0.12

Cl as ppm; F, S and CO_2 as wt.%

	SP835	SP836	SP838	SP839	SP842	SP845
Au*	17	19	19	17	12	17
Au#	18	–	–	14	–	–
Pd#	8	–	5	12	2	4
Pt*	44	52	79	88	42	48
Pt#	20	–	–	10	–	–
Ru*	20	17	18	19	17	18
Ir*	1.5	1.4	1.2	0.3	0.7	0.7
Os*	3.5	–	3.1	3.4	3.1	3.8
Ag*	115	81	87	96	84	91
Hg* ppm	12.6	10.6	11.1	11.6	10.2	11

* = INAA
# = Fire assay with Pb and AAS finish
The results are in ppb, except where otherwise stated
– = not determined.

Gold with these characteristics, in lamproite magmas, has not been found previously.

Au and PGE contents of rock

The Au and PGE contents, determined from the rocks of El Tale are reported in Table 2. The scarcity of Au and PGE data for the Spanish lamproites prevents any comparison and makes it impossible to relate mineralogy and bulk rock composition to Au and PGE chemistry, at the scale of the single outcrop. Whole-rock Au and PGE analyses have been performed using different techniques: (1) Au, Pt and Pd: Pb-fire assay procedure with Atomic Absorption finish (30 g of sample; detection limits 1 ppb for Au and Pd and 10 ppb for Pt); (2) Au, Pt, Rh, Ru, Ir and Os: INAA (1 g of sample; detection limit 5 parts in 10^{12}, University of Pavia). Rhodium is below the detection limit.

The mantle normalized element patterns are reported in Fig. 3. Gold displays a broad positive anomaly and is positively related to Pt, whereas Ir displays a negative anomaly. The Au, Pd, Pt and Ru are higher than in the primitive mantle (Barnes *et al.*, 1988); this enrichment could be related to the composition of the mantle sources or to the fractionation of the lamproite magma during the way toward the surface. Effectively, the negative anomaly of Ni and the high Cu/Ni (0.23) ratio may be explained by fractionation of olivine. Platinum-group elements are fractionated (Pd/Ir = 2.9–120); the Pd/Au (0.42–0.86) ratio is much lower than in the primitive mantle (3.64; Brugmann *et al.*, 1987).

Discussion

The gold grains found in the lamproite of El Tale can be considered to be of magmatic origin on the basis of the following considerations: (1) Metal spheroids similar to those found at El Tale have been crystallized during the experiments on the solubility of noble metals in basic silicate melts (Amossé *et al.*, 1990; Amossé and Allibert, 1993). (2) The freshness of the volcanic rock, especially of the glassy groundmass containing the gold grains, the absence of an apparent enrichment of Au grains along the wall and in the most altered portions of the dyke, and the low contents of Au 'path-finder' elements such as As, Sb, S and Cl exclude hydrothermal input of gold. (3) Mann (1984)

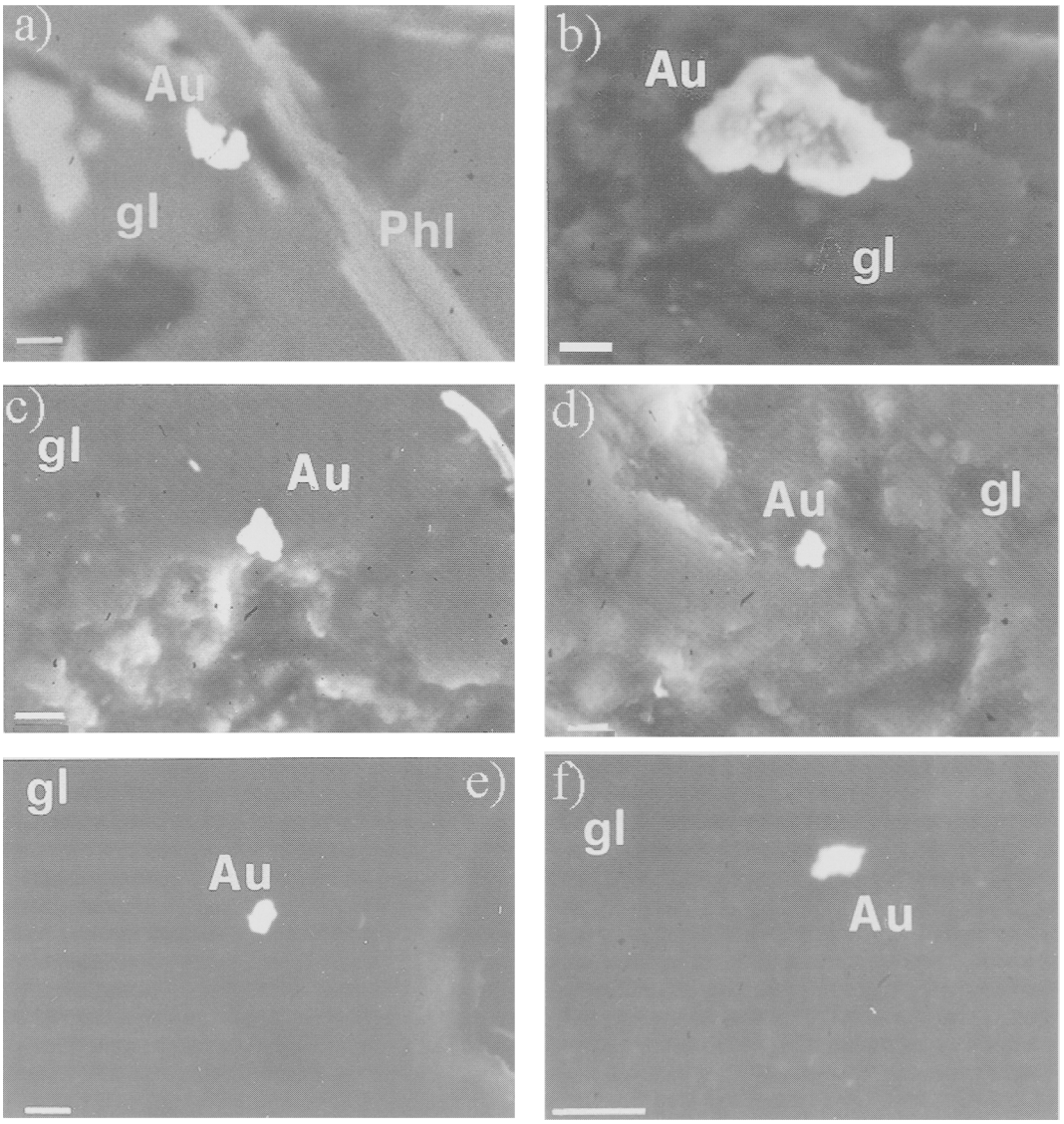

FIG. 2. SEM images: Au = gold grains, Phl = phlogopite, gl = glass. Scale bars = 1 μm.

reports the occurrence of gold grains (small octahedral crystals and sponge gold) of high purity (Ag <0.4 wt.%) in lateritic weathering profiles from Western Australia and relates the origin of the grains to preferential leaching of Ag from pre-existing Ag-bearing gold nuggets. In our case, however, the freshness of the lamproitic rock (point 2) excludes any significant leaching processes. (4) The following arguments support a sub-crustal origin for gold: (a) in the magma of El Tale, contamination by crustal material is quantitatively insignificant (rare xenocrysts of biotite having Al_2O_3 contents of ~20 wt.%, and the Al_2O_3 enrichment of the late-crystallizing dark mica and clinopyroxene; Toscani and Salvioli-Mariani, submitted); (b) gold deposits in the sedimentary and metamorphic formations of the wall rocks are unknown at the regional scale; and (c) the Au content found at El Tale is greater than in the average crust (Au = 3 ppb, Taylor and McLennan, 1985).

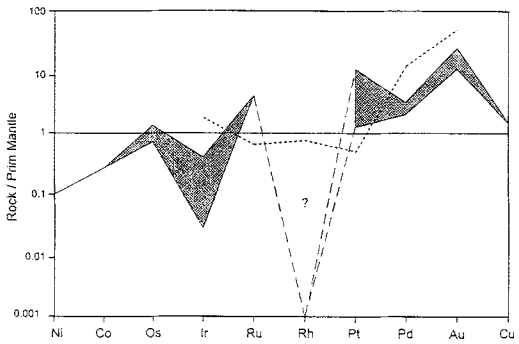

FIG. 3. Mantle normalized abundances of Group VIII metals plus Cu and Au. Greyish field = El Tale lamproite, dotted line = Ellendale-11 lamproite (West Kimberley, Western Australia; Lewis, 1987). Primitive mantle normalizing values are from Barnes *et al.*, (1988).

Speciation, transport and fractionation of Au and noble metals in magmas are not well known, and a detailed discussion on the mechanism of Au (and PGE) concentration in the lamproite of El Tale is beyond the aim of this paper, but some observations can be made. Barnes *et al.* (1988, and references therein) have suggested an important role for olivine and chromite in Au and PGE fractionation. According to these authors, olivine and chromite cumulates are Os-Ir-Ru-rich and Pt-Pd-Rh-Au-poor, although it is a matter of debate whether Os, Ir and Ru enter the cumulate minerals as platinum-group minerals (PGM, Barnes *et al.*, 1985; Davies and Tredoux, 1985) or by solid substitution (Brugman *et al.*, 1985). On the other hand, the experiments of Amossé *et al.* (1987, 1990) and Amossé and Allibert (1993) show that Au and PPGE (Pt, Pd, Rh) are more soluble than IPGE (Os, Ir, Ru) in basaltic liquids, and Keays (1982) determined the contents of Pd and Ir in both olivine and its host dunitic komatiite showing that the olivine/liquid partition coefficients are lower than unity ($K_{Pd} < K_{Ir} < 1$), suggesting an incompatible behaviour for all of the PGE. These results indicate that olivine ($\pm$ chromite) segregation may lead to an overall fractionation of Au and PGE as shown in Fig. 3 for the rocks of El Tale, which underwent ~10% olivine fractionation (Toscani and Salvioli-Mariani, submitted). Other features such as: (1) the saw-tooth edged distribution of the IPGE; and (2) the abundances of Ru and Os, which are greater than in the primary mantle, cannot be explained simply. In any case, points (1) and (2), the contents of Au, Pd, Pt being greater than in the primitive mantle, and the sulfur content of the rock, seem to exclude the fractionation of these elements in a sulfide liquid phase during partial melting in the mantle sources. The lamproite from Ellendale-11 (West Kimberley, Western Australia; Lewis, 1987) differs from the rocks of El Tale in having high Au-Pd-Ir and low Pt-Rh-Ru (Fig. 3). The different distribution may be related to the different evolution of the mantle under the Betic Cordillera and the Kimberley Block (Nixon *et al.*, 1984; Foley, 1989, 1992; Venturelli *et al.*, 1988).

Amossé *et al.* (1990) and Amossé and Allibert (1993) found that the solubility of Au and PGE is inversely related to f_{O_2} in the range of values $10^{-4}-10^{-7}$ atm (QFM minus 1–2 log unit), and Amossé and Allibert (1993) infer that Au and PGE enter the silicate liquid as zerovalent atoms at low oxygen fugacity ($f_{O_2} = 10^{-4}-10^{-7}$ atm $\approx$ QFM minus 1–2 log unit). It is noteworthy that the redox conditions of the lamproitic magma of El Tale ($f_{O_2} \approx$ IW) are similar to those which are compatible with the highest metal solubility in the experiments of Amossé and Alibert (1993). The hypothesis of a zerovalent state of Au in the silicate liquid is in agreement with the occurrence of gold grains in the glassy groundmass of El Tale lamproite. The high contents of Pd, Pt and Ru (and Os), however, are difficult to explain by a zerovalent state of these elements, because of their high melting temperature as metals (Pd = 1552°, Pt = 1772°, Ru = 2310°C). Since Au and PGE form halide- and alkali-halide-complexes (Cotton, 1997), having relatively low temperatures of crystallization, the complexation of the noble metals in mafic melts may be invoked. For instance, the role of F as a noble metal carrier during the magmatic processes has been suggested by Rock and Groves (1988) after the experiments of Letnikov and Vilor (1981) on the high-temperature stability of gold-fluoride complexes. Actually, the lamproite magma of El Tale exhibits high K and F contents, and thus solubility of the noble metals as fluoride- and alkali-fluoride-complexes is a realistic hypothesis.

Acknowledgements

This study was supported by MURST 40% and CNR-Roma (grant 97.00295.CT05: G.

Venturelli), MURST 60% (L. Toscani) and by COFIN-MURST-97 (G. Serri). I am grateful to Prof. M. Oddone (Pavia), who performed the noble metals determinations by INAA; to Professors G. Garuti (Modena), P. Lattanzi (Firenze), and G. Venturelli (Parma); to the referees, Prof. Steve Barnes, Dr John Bowles and another anonymous referee, for useful suggestions and constructive review of the manuscript; to Dr E. Salvioli-Mariani (Parma) for her help with the SEM-EDS analyses; and finally to Mrs E. Masini for drawing Figs 1 and 3. The microprobe analyses (Jeol 6400 with Tracor EDS) and XRF analyses (Philips PW1400) were performed at the Department of Earth Sciences of Parma University.

References

Amossé, J. and Allibert, M. (1993) Partitioning of iridium and platinum between metals and silicate melts: evidence for passivation of the metals depending on f_{O_2}. *Geochim. Cosmochim. Acta*, **57**, 2395–8.

Amossé, J., Allibert, M., Fischer, W. and Piboule, M. (1987) Étude de l'influence des fugacities d'oxigene et de soufre sur la differenciation des platinoides dans les magmas ultramafiques. Resultats preliminareires. *C. R. Acad. Sci. Paris*, t304 Seri II, **19**, 1183–5.

Amossé, J., Allibert, M., Fischer, W. and Piboule, M. (1990) Experimental study of the solubility of platinum and iridium in basic silicate melts – Implications for the differentiation of platinum-group elements during magmatic processes. *Chem. Geol.*, **81**, 45–53.

Barnes, S.J., Naldrett, A.J. and Gorton, M.P. (1985) The origin and the fractionation of platinum-group elements in terrestrial magmas. *Chem. Geol.*, **53**, 303–23.

Barnes, S.J., Boyd, R., Korneliusson, A., Nilsson, L.P., Often, M., Pedersen, R.B. and Robins, B. (1988) The use of mantle normalization and metal ratios in discriminating between the effects of partial melting, crystal fractionation and sulfide segregation on platinum-group elements, gold, nickel and copper: examples from Norway. In *Geo-platinum '87* (H.M. Prichard, P.J. Potts, J.F.W. Bowles and S.J. Cribb, eds). Elsevier, 113–43.

Brugmann, G.E., Arndt, N.T., Hofmann, A.W. and Tobschall, H.J. (1985) Precious-metal abundances in komatiites and komatiitic basalts: implications for the genesis of PGE-bearing magmatic sulfide deposits. *Canad. Mineral.*, **23**, 293–321.

Brugmann, G.E., Arndt, N.T., Hofmann, A.W. and Tobschall, H.J. (1987) Noble metal abundances in komatiite suites from Alexo, Ontario, and Gorgona Island, Colombia. *Geochim. Cosmochim. Acta*, **51**, 2159–69.

Contini, S., Venturelli, G., Toscani, L., Capedri, S. and Barbieri, M. (1993) Cr-Zr-armalcolite-bearing lamproites of Cancarix, SE Spain. *Mineral. Mag.*, **57**, 203–16.

Cotton, S.A. (1997) *Chemistry of precious metals*. Blackie Academic and Professional, 374 pp.

Davies, G. and Tredoux, M. (1985) The platinum-group element and gold contents of the marginal rocks and sills of the Bushveld Complex. *Econ. Geol.*, **80**, 838–48.

De Larouzière, F.D., Bolze, J., Bordet, P., Hernandez, J., Montenat, C. and Ott d'Estevou, P. (1988) The Betic segment of the lithospheric Trans-Alboran shear zone during the Late Miocene. *Tectonophysics*, **152**, 41–52.

Foley, S.F. (1989) The genesis of lamproitic magmas in a reduced fluorine–rich mantle. In *Kimberlites and related rocks*, Vol 1. (A.L. Jacques, J. Ferguson and D.H. Green, eds). Blackwells, pp. 616–31.

Foley, S.F. (1992) Vein-plus-wall-rock melting mechanisms in the lithosphere and the origin of potassic alkaline magmas. *Lithos*, **28**, 435–53.

Foley, S.F., Venturelli, G., Green, D.H. and Toscani, L. (1987) The ultrapotassic rocks: characteristics, classification and constraints for petrogenetic models. *Earth Sci. Rev.*, **24**, 81–134.

Fuster, J.M., Gastesi, P., Sagredo, J. and Fermoso, M.L. (1967) Las rocas lamproiticas del SE de España. *Est. Geol. Madrid*, **23**, 35–69.

Jaques, A.L. (1990) Do lamprophyres have high precious-metal contents? *Bur. Miner. Resources Res. Newsl.*, **12**, 17–8.

Jaques, A.L., Lewis, J.D. and Smith, C.B. (1986) The kimberlites and lamproites of Western Australia. *Geol. Surv. W. Australia Bull.*, **132**, 268 pp.

Keays, R.R. (1982) Palladium and iridium in komatiites and associated rocks: application to petrogenetic problems. In *Komatiites* (N.T. Arndt and E.G. Nisbet, eds). Allen, pp. 435–57.

Kerrich, R. and Wyman, D.A. (1994) The mesothermal gold-lamprophyre association: significance for an accretionary geodynamic setting, supercontinent cycles, and metallogenic processes. *Mineral. Petrol.*, **51**, 147–72.

Letnikov, F.A. and Vilor, N.V. (1981) Gold in the hydrothermal process. *Nauka* (in Russian), Moscow, 224 pp.

Lewis, J.D. (1987) *The geology and geochemistry of the West Kimberley lamproite province, Western Australia*. M.Sc. thesis, Univ. Western Australia, Perth.

Mann, A.W. (1984) Mobility of gold and silver in

lateritic weathering profiles: some observations from Western Australia. *Econ. Geol.*, **79**, 38–49.

Nixon, P.H., Thirlwall, M.F., Buckley, F. and Davies, C.J. (1984) Spanish and western Australian lamproites aspects of whole rock geochemistry. In *Kimberlites and related rocks* (J. Kornprobst, ed.). Elsevier, pp. 285–96.

Rock, N.M.S. and Groves, D.I. (1988) Can lamprophyres resolve the genetic controversy over mesothermal gold deposits? *Geology*, **16**, 538–41.

Sekerin, A.P., Men'shagin, Yu.V. and Lashchenov, V.A. (1993) Sayan Precambrian lamproites. *Trans. Russ. Acad.* (Dokl. Acad. Nauk 1993, 329/3,328–331), **329A**, 99–104.

Taylor, S.R. and McLennan, S.M. (1985) *The continental crust: its composition and evolution.* Blackwell Scientific Publications, 312 pp.

Taylor, W.R., Rock, N.M.S., Groves, D.I., Perring, C.S. and Golding, S.D. (1994) Geochemistry of Archean shoshonitic lamprophyres from the Yilgarn Block, Western Australia: Au abundance and association with gold mineralization. *Appl. Geochem.*, **9**, 197–222.

Toscani, L. and Salvioli-Mariani, E. The lamproite of El Tale (Fortuna, Southeast Spain). Submitted.

Venturelli, G., Capedri, S., Di Battistini, G., Crawford, A., Kogarko, L.N. and Celestini, S. (1984) The ultrapotassic rocks from southeastern Spain. *Lithos*, **17**, 37–54.

Venturelli, G., Salvioli-Mariani, E., Foley, S.F., Capedri, S. and Crawford, A. (1988) Petrogenesis and conditions of crystallization of Spanish lamproitic rocks. *Canad. Mineral.*, **26**, 67–79.

[*Manuscript received 10 January 1999: revised 7 June 1999*]