A sapphirine–cordierite–bronzite–phlogopite paragenesis from Namaqualand, South Africa

T. N. CLIFFORD,[1] E. F. STUMPFL,[2] AND J. R. McIVER[1]

SUMMARY. The rock composition and electron microprobe analyses of the five constituent minerals are presented for a sapphirine–cordierite–bronzite–phlogopite–spinel rock from Namaqualand, South Africa. These data are considered in relation to experimental data and the mineralogy of certain associated rock types, and it is suggested that the sapphirine-bearing rock represents a metamorphic residuum (restite) after extraction of anatectic granitic liquid from an original argillaceous sediment.

THE major part of Namaqualand is underlain by a sequence of gneisses and granulites of granitic composition, with quartzite and schist and a number of more local rock-types including hornblende and hypersthene gneiss and granulite. Recognizable meta-sedimentary and metavolcanic rocks are usually ascribed to the Kheis System (Martin, 1965, p. 59).

In the Nababeep district the structural succession comprises (Benedict et al., 1964): Brandberg gneiss; Springbok granulite, quartzite, and schist; Nababeep gneiss; Concordia granite and gneiss with Wolfram schist; and Ratelpoort granulite, quartzite, and schist with augen gneiss. Gneiss and granulite from this suite have yielded a Rb-Sr whole-rock isochron age of 1213±22 Myr that is considered to reflect the main regional metamorphism (Clifford et al., 1975).

The metamorphic pattern of Namaqualand has been interpreted in terms of a regional thermal dome (Joubert, 1971, p. 123). The Nababeep district lies on the western side of its culmination and the grade of metamorphism is illustrated by the absence of primary muscovite, and by the presence of sillimanite–garnet–microcline microperthite, sillimanite–cordierite, and hornblende–hypersthene–clinopyroxene assemblages in rocks of appropriate composition. In addition, some years ago, the geologists of the O'okiep Copper Company recorded sapphirine-bearing rocks in the Springbok schist (Venter, 1951); and it is the purpose of this paper to present the results of an investigation of an example of one of these parageneses from Nababeep Kloof.[3] At that locality, sapphirine rock occurs as three concordant bodies within a northward-dipping succession of interbedded schist and quartzite. The example described here is the most northerly of the bodies. It has a strike length of 30 m and a maximum thickness of 1·5 m and, whilst its 'downdip' dimension is not known, it is believed that all three occurrences represent a single horizon repeated by folding; on this reasoning the 'downdip' dimension is not less than 200 m.

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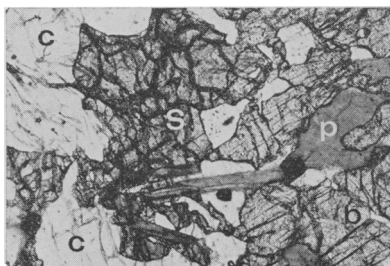[3] The locality (no. 32) is shown on a geological map in Clifford et al. (1975)

FIG. I. Photomicrograph of the sapphirine(s)–cordierite(c)–bronzite(b)–phlogopite(p) rock, Nababeep Kloof. ×22.

TABLE I. *Electron-probe analyses* and optical properties† of minerals from sapphirine–bronzite–cordierite–phlogopite rock*

	Sapphirine‡	Bronzite	Cordierite	Phlogopite	Spinel
SiO_2	14·1	53·2	50·1	38·5	—
TiO_2	—	—	—	3·9	—
Al_2O_3	61·6	7·1	34·9	16·4	66·4
FeO	4·9	11·5	1·3	4·9	14·3
MgO	19·8	27·9	13·6	20·6	19·7
K_2O	—	—	—	12·0	—
H_2O	—	—	—	4·0	—
	100·4	99·7	99·9	100·3	100·4

Number of ions

N(O,OH)	10(O)	6(O)	18(O)	24(O,OH)	4(O)
Si	0·827 } 1·00	1·876 } 2·00	4·922 } 6·00	5·512 } 8·00	—
Al	0·173 ⌡ ; 4·00 ; 0·079 ⌐	0·124 ⌡ ; 0·172 ⌐	1·078 ⌡ ; 2·959 } 3·00	2·488 ⌡ ; 0·281 ⌐	1·972 } 2·00
Fe	0·239 } 2·05	0·339 } 1·98	0·041 ⌡ ; 0·065 } 2·06	0·585 } 5·68	0·028 ⌡ ; 0·273 } 1·01
Mg	1·728 ⌡	1·465 ⌡	1·995 ⌡	4·394	0·741 ⌡
Ti	—	—	—	0·42 ⌡	
K	—	—	—	2·18	
OH	—	—	—	3·98	—

$\dfrac{Fe}{Fe+Mg}$	0·12	0·19	0·05	0·12	0·29

Optical properties

α	1·709	1·662	1·536	n.d.	
β	1·716	1·667	1·543	1·597	
γ	1·724	1·685	1·547	1·601	
$2V_\alpha$	65°	96°	92–96°	0°	

* *Analyst*, E. F. Stumpfl. Analyses were made using an ARL EMX–SM microprobe; the results were processed by the computer programme EMPADR (Rucklidge, 1967). Total Fe is given as FeO.

† R.I. determinations were carried out on a universal stage using Na light, and immersion liquids were calibrated using an Abbé refractometer; the R.I. values are reported to 0·002. 2V values were obtained by conventional orthoscopic universal-stage measurements.

‡ Venter (1951) has given an analysis of sapphirine, probably from the present locality: SiO_2, 14·56%; TiO_2, 0·35%; Al_2O_3, 60·17%; Fe_2O_3, 2·97%; FeO, 3·59%; MgO, 17·22%; Na_2O, 0·10%; K_2O, 0·05%; H_2O+, 0·64%; H_2O-, 0·20%: Total 99·87%. Analyst, C. F. J. van der Walt.

The sapphirine rock is essentially composed of cordierite, phlogopite, bronzite, and sapphirine (fig. 1); electron microprobe analyses of these minerals and of accessory spinel are given in Table I. *Phlogopite* containing 3·9 % TiO_2 occurs as randomly oriented crystals with α, colourless/pale yellow brown; β, γ, yellow brown. *Cordierite* forms aggregates of polysynthetically twinned crystals that are readily recognized by the local presence of clusters of ramifying fractures typically occupied by a micaceous alteration product (fig. 1). Optically the crystals exhibit some variation in $2V_\alpha$ from 96° in the core to 92° in the rims. The ortho-pyroxene is an *aluminous bronzite* containing 7·1 % Al_2O_3. Locally it forms up to 50 % of the rock and contains inclusions of cordierite and phlogopite; the presence of abundant optically continuous, but physically separated, inclusions of the latter suggests that bronzite developed partly at the expense of phlogopite. *Sapphirine* is sited largely within cordierite granular aggregates but is also locally concentrated along the margins of crystals of aluminous bronzite (fig. 2*a*). It has α, colourless/pale pink; β, pale blue; and γ, blue-green. It shows irregular fractures with a brown unidentified alteration product. Phlogopite, cordierite, bronzite, and spinel occur as inclusions and there is convincing petrographic evidence that sapphirine developed, in part, from phlogopite (fig. 2*b*). *Spinel* with associated *carbonate* and *rutile* are present in accessory amounts. Of these, the green spinel is *ceylonite* with an $Fe/(Fe+Mg)$ ratio of 0·29. From its ubiquitous presence as inclusions in sapphirine, and thus armoured from the surrounding cordierite, it is considered that the pair cordierite+spinel was unstable.

This rock shows earlier-formed cordierite and phlogopite, and later-formed aluminous bronzite and sapphirine. In this regard, Seifert
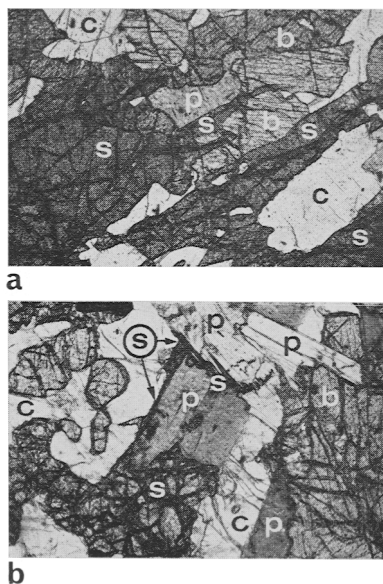

Fig. 2. (*a*) Photomicrograph showing the relationship between bronzite(b) and sapphirine(s); note the tongue-like sapphirine in bronzite to the northeast of the central s. Other minerals are cordierite (c) and phlogopite (p). ×22. (*b*) Photomicrograph of replacement of phlogopite (p) by sapphirine (s) locally developed at cordierite-phlogopite boundary. ×22.

(1974) has recently established the appearance of a stable tie-line enstatite+sapphirine above 3·5 Kb and at temperatures in excess of 765° C (curve 1, fig. 3); these data provide an *estimate* of the minimum P and T of formation of the Nababeep bronzite +sapphirine. That significantly higher temperatures were attained is suggested by the alumina content of the bronzite, because Anastasiou and Seifert (1972) have shown that, in an alumina-saturated environment, the Al-content of orthopyroxene is rather a sensitive geothermometer; the solvus for enstatite containing 7·0 % Al_2O_3 (curve 2, fig. 3) is appropriate to the Nababeep paragenesis and suggests that tempera-tures close to 1000 °C may have been attained. In an attempt to define the minimum

pressure of metamorphism with greater precision, garnets from a number of horizons in the Nababeep area have been analysed using the electron microprobe (Table II). Of these, garnet No. 10a is an unsuitable PT indicator because of its high spessartine content. However, utilizing the curves for the stability of pyrope and almandine determined by Schreyer (1968) and Keesmann et al. (1971) respectively, the 'crude' approximation to the minimum stability of $Alm_{80}Py_{20}$ (see fig. 3) gives some measure of
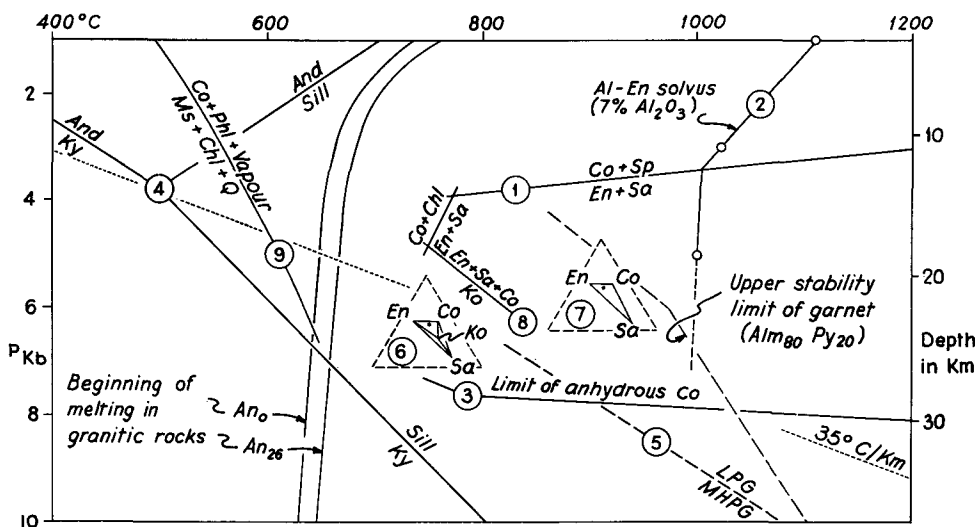

FIG. 3. Some experimental data relevant to the discussion of the petrogenesis of the Nababeep sapphirine-bearing rock. *Abbreviations*: Al-En, aluminous enstatite; Alm, almandine; And, andalusite; Chl, chlorite; Co, cordierite; En, enstatite; Ko, kornerupine; Ky, Kyanite; Ms, muscovite; Phl, phlogopite; Py, pyrope; Q, quartz; Sa, sapphirine; Sill, sillimanite; and Sp, spinel. *Sources of data*: 1, Seifert (1974); 2, Anastasiou and Seifert (1972); 3, Newton (1972); 4, Holdaway (1971); 5, boundary between low-pressure granulite (LPG) and medium-to-high pressure granulite (MHPG), from Green and Ringwood (1967); 6–8, Seifert (1975); and 9, Seifert (1970). For the chemical components of diagrams 6 and 7 see fig. 4. The curves for the beginning of melting of rocks of granitic composition are: from Luth et al. (1964), An_0; and from Winkler (1970), An_{26}. Upper stability limit of $Alm_{80}Py_{20}$ (stable to the left) is calculated from experimental data given by Schreyer (1968) and Keesmann et al. (1971).

the minimum pressure limit of garnets nos. 10b, 11 and 13. When considered in relation to the Al-enstatite solvus (curve 2, fig. 3) a minimum pressure of 6·0 Kb is suggested for the metamorphism in this region. Excessively high pressures are, however, precluded because, even with $P_{H_2O} = P_{total}$ at the time of metamorphism, the continued presence of cordierite and cordierite+sapphirine rather than enstatite+sillimanite in the Nababeep sapphirine rock indicates that pressures of c. 10–11 Kb were not exceeded (Schreyer and Yoder, 1964; Schreyer and Seifert, 1969, p. 378). If, moreover, the system was anhydrous, a maximum pressure of 7·5–8·0 Kb is implied by Newton's (1972) experimental data on the reaction cordierite = sapphirine+quartz+(enstatite?) (see curve 3, fig. 3).

Cumulatively, these data are consistent with: the observed regional path of prograde metamorphism via kyanite- (not andalusite-)bearing assemblages (Joubert, 1971), which suggests an *average* geothermal gradient of *less* than 35 °C/Km (see curves 4, fig. 3); and the presence, in the Nababeep gneiss, of rocks of basic composition, including olivine-normative types, that are characterized by the assemblage hypersthene (after hornblende)+clinopyroxene+plagioclase, which Green and Ringwood (1967) consider to be typical of medium-to-high pressure granulites (curve 5, fig. 3).

TABLE II. *Compositions* of garnets from some Nababeep rocks*

Sample no.†	Rock type	Major garnet end-members ($> 5\%$)
10a	Concordia gneiss	$Almandine_{73}Spessartine_{12}Pyrope_{12}$
10b	Wolfram schist	$Almandine_{67}Pyrope_{25}$
11	Springbok granulite	$Almandine_{71}Pyrope_{18}Spessartine_{7}$
13	Springbok granulite	$Almandine_{81}Pyrope_{11}$

* Electron microprobe analyses by E. F. Stumpfl.
† See Clifford *et al.* (1975) for localities.

The data imply, moreover, that kornerupine assemblages, particularly kornerupine+enstatite+cordierite (see 6, 7, and 8 of fig. 3; from Seifert, in manuscript) or kornerupine+aluminous anthophyllite+enstatite (see fig. 4; Yoder, 1971) may be expected at lower temperatures in rocks of bulk composition similar to those of the Nababeep sapphirine rock.

Parageneses similar to the Nababeep sapphirine rock have been recorded from Greenland (Herd *et al.*, 1969), Australia (Hudson and Wilson, 1966), Italy (Barker, 1964), Norway (Touret and De la Roche, 1971), Russia (Chekirda and Entin, 1969), Antarctica (Segnit, 1957), India (Muthuswami, 1949), Brazil (Fujimori and Allard, 1966), and Rhodesia (Robertson, 1973). A variety of origins have been proposed of which three are particularly important: metasomatism of mafic complexes (Chekirda and Entin, 1969; Foissy *et al.*, 1966) or magnesium metasomatism of pelites (Muthuswami, 1949); recrystallization of ultramafic rocks under hornblende granulite subfacies or cordierite amphibolite subfacies metamorphism (Deer *et al.*, 1962a, pp. 179–81; Lensch, 1971) with accompanying Si, Ca, and K metasomatism (Herd *et al.*, 1969); and granulite facies metamorphism of magnesian argillites (Segnit, 1957; Touret and De la Roche, 1971).

The chemical composition of the Nababeep sapphirine rock[1] given in Table III(a) shows that it is largely composed of MgO, Al_2O_3, and SiO_2. A hypothesis based on isochemical metamorphism of a former basic or ultrabasic rock cannot be reconciled with the high alumina and the virtual absence of Ca and Na. K metasomatism is

[1] It is of interest to note that the composition of this sapphirine rock is very similar to that of pyrope-rich garnet. The recalculated rock chemistry yields the theoretical assemblage 92·2 % garnet ($Py_{86.7}Alm_{12.8}Gr_{0.3}Sp_{0.2}$) with small amounts of enstatite (2·2 %), ilmenite (1·1 %), magnetite (0·9 %), Fe_2O_3 (1·0 %) and small amounts of K_2O, Na_2O, P_2O_5, and H_2O.
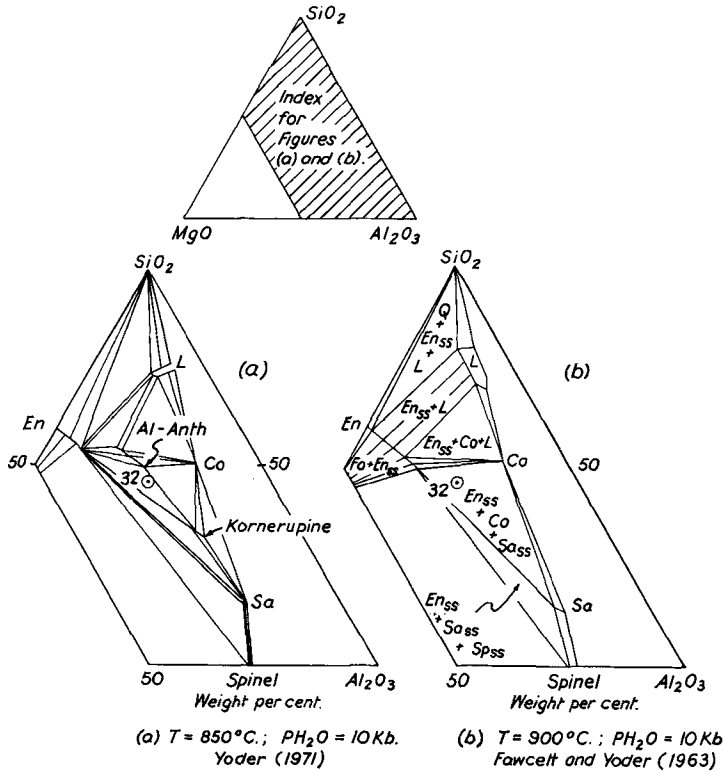
FIG. 4. Nababeep sapphirine rock (no. 32) plotted in relation to isothermal sections through the system MgO–Al₂O₃–SiO₂–H₂O. *Abbreviations* as in fig. 3 together with: Al-Anth, aluminous anthophyllite; Fo, forsterite; L, liquid; and SS, solid solution.

ruled out because of the evidence for the *loss* of potassium during the development of sapphirine and bronzite presumably reflecting a reaction of the type:

4 phlogopite+2 cordierite → 2 sapphirine+3 bronzite+rutile+MgO,

with the concomitant loss of K_2O, Al_2O_3, SiO_2, and H_2O equivalent to 4K-feldspar+ $4H_2O$ from the system. Furthermore, the development of the sapphirine rock-composition by massive Mg-metasomatism of a former sediment or acidic-intermediate volcanic rock or Al-metasomatism of an ultramafic rock is not favoured; the former because of the complex chemical losses that would have to be invoked, and the latter because no ultramafic source material is discernible in the field. The association of the sapphirine rock with the Springbok schist of pelitic and semipelitic composition favours an origin from argillaceous sediments, as suggested by Venter (1951).

It has been noted that the Nababeep sapphirine rock records an earlier cordierite–phlogopite assemblage and a later Al-bronzite+sapphirine paragenesis. In this regard Seifert (1970) has presented experimental data for the reaction (curve 9, fig. 3):
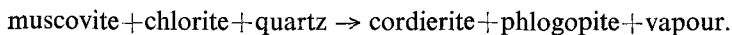
muscovite+chlorite+quartz → cordierite+phlogopite+vapour.

TABLE III. a–c, *composition of the Nababeep rock compared with certain theoretical and natural compositions*; a'–f', *computed best-fit compositions of mixtures of sapphirine rock and alkali granite compared with average pelite and sandy argillite*

	a	b	c		a'	d	e	e'	f	f'
SiO_2	41·50	37·45	37·82		42·18	74·20	64·10	70·08	67·51	66·33
TiO_2	0·36	—	0·22		0·37	0·20	0·86	0·22	0·50	0·24
Al_2O_3	22·71	22·24	14·72		23·08	13·81	17·70	15·00	12·53	16·09
Fe_2O_3	2·33	1·88	6·47		2·37	0·78	2·70	0·98	2·24	1·17
FeO	5·38	2·75	8·90		5·47	1·14	4·05	1·70	3·72	2·20
MnO	0·08	—	0·37		—	—	—	—	—	—
MgO	24·87	23·05	14·36		25·29	0·26	2·65	3·48	5·49	6·41
CaO	0·09	—	1·81		0·09	0·72	1·88	0·64	0·76	0·57
Na_2O	0·06	0·15	1·10		0·06	3·53	1·91	3·08	2·56	2·68
K_2O	0·85	2·73	1·10		0·86	5·15	3·60	4·60	2·91	4·10
H_2O+	1·21	9·80	7·10							
H_2O-	0·13	—	6·44							
P_2O_5	0·05	—	—							
	99·62	100·05	100·41							

a. Cordierite–sapphirine–bronzite–phlogopite rock, Nababeep Kloof, South Africa. Analyst, E. Clementson.

b. Composition of chlorite–muscovite schist calculated for a mineral composition of 6chl+3ms+4qz based on mineral analyses for muscovite (Deer *et al.*, 1962*b*, p. 16, anal. 1) and chlorite (ibid., p. 143, anal. 26)

c. Interstratified chlorite–montmorillonite alteration product of tuff and tuffaceous sediment, Yamakata-machi, Japan. (Kimbara *et al.*, 1971).

a'. Sapphirine rock, a, recalculated anhydrous; excluding MnO, and P_2O_5.

d. Average alkali granite (Nockolds, 1954, p. 1012), recalculated anhydrous; excluding MnO, P_2O_5, and other minor components.

e. Average pelite (Shaw, 1956, p. 928), recalculated anhydrous; excluding MnO, P_2O_5, and other minor components.

e'. Computed composition of (a'+d) that best fits the composition of average pelite (e), based on 12·9 % a'+87·1 % d (D-normalization*).

f. Sandy argillite, Lomagundi district, Rhodesia (Phaup, 1973, p. 194), recalculated anhydrous; excluding MnO, P_2O_5, and other minor components.

f'. Computed composition of (a'+d) that best fits the composition of sandy argillite (f), based on 24·6 % a'+75·4 % d (D-normalization*).

* Normalization by the dependent vector; all data for the mix divided through, element by element, by the elemental abundances in the dependent vector (e for e', f for f').

Viewed in these terms, the sapphirine rock can be recalculated most simply to 6 clinochlore+3 muscovite+4 quartz. For the purposes of comparison, Table III(*b*) shows the composition of a rock with these mineral proportions, calculated from clinochlore and muscovite analyses given by Deer *et al.* (1962*b*). Clearly neither this hypothetical rock nor the sapphirine-bearing rock corresponds to any commonly occurring igneous or sedimentary rock. It is, of course, just possible that the pre-metamorphic rock was a smectite(saponite)-rich bentonite (Ross and Hendricks, 1945) or a chlorite–montmorillonite mixture similar to that which occurs as an alteration product of tuffs and tuffaceous sediments in Japan (Kimbara *et al.*, 1971; see Table III(*c*)). However, although such an origin is feasible, it ignores the influence of metamorphism in promoting bulk-chemical change in the Nababeep district.

In this regard it is of some importance to note that Wyllie and Tuttle (1961) showed that with $P_{H_2O} = P_{total}$ shales begin to melt at temperatures not greatly in excess of the melting temperature of granite (see fig. 3); and that 150 °C above the temperature of melting the fused shales consist of 50 % of liquid of granite–granodiorite composition together with crystals of *cordierite, hypersthene,* quartz, and mullite; such a temperature condition was apparently attained during metamorphism in the Nababeep district (see fig. 3). In similar studies, James and Hamilton (1969) noted that clay produced 63 % of melt of alkali feldspar+quartz composition at 810 °C and 1 Kb. To test the plausibility of such a process in the development of the Nababeep sapphirine rock, the 'Mixer' computer programme written by Dr. A. R. Duncan (University of Cape Town) has been used to determine the percentages of sapphirine rock (Table III(a')) and alkali granite (Table III(d)) required to provide compositions that best fit those of average pelite (Table III(e)) and of sandy argillite (Table III(f)). These data provide a reasonable comparison between computed mixtures of 25 % sapphirine rock plus 75 % alkali granite (Table III(f')) and sandy argillite (Table III(f)). Moreover, bearing in mind the large standard deviations given by Shaw (1956, p. 928) for all components of the average pelite (Table III(e)), the apparent poor correlation between the composition of the latter and the best fit mix of sapphirine rock plus alkali granite (Table III(e')) cannot be regarded as significant. In view of its field association with the Springbok schist of pelitic and semipelitic composition, and in the light of the *PT* conditions of Nababeep metamorphism and the results of experimental melting on argillaceous rocks, we interpret the Nababeep sapphirine rock as a metamorphic residuum (restite) after the extraction of 75–80 % of anatectic granitic liquid from an argillaceous sediment.[1] The dimensions of the sapphirine bodies, given earlier in this paper, suggest that the actual volume of magma developed from this source was small. Moreover, since sapphirine could not have coexisted with alkali granite at the pressures invoked for the metamorphism (H. S. Yoder, personal communication), it follows that the granitic liquid was lost from the system at temperatures lower than sapphirine stability. Because of the widespread nature and variety of rocks of granitic composition in the Nababeep region, it must be uncertain what became of this expelled liquid.

[1] It is worthy of note that Leake and Skirrow (1960) have suggested a similar type of restite origin for xenoliths of spinel+magnetite+cordierite±orthopyroxene±corundum parageneses in Eire and suggest that the composition of the material removed from the xenoliths resembled that of subalkaline granite. In addition, Grant (1968) has argued that certain cordierite–anthophyllite rocks may represent a residuum after the extraction of a granitic melt from certain rock types, especially metamorphosed sediments of greywacke composition. Cordierite–anthophyllite–hypersthene rocks that have been recognised by F. J. G. Schreuder in interbedded schist and quartzite on Farm Goinoep, to the south-east of Nababeep, will be the subject of a future communication.

through grant no. Stu. 77/1 to E. F. Stumpfl; Miss C. Dettmann for her help with the probe analyses; Mrs. E. Clementson for the analysis of the sapphirine rock; and Mrs. S. McCarthy and Mr. M. Hudson for secretarial and photographic services respectively.

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