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The geochemistry of 'charnockites' and their constituent ferromagnesian minerals from the Precambrian of south-east Sri Lanka (Ceylon)

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SUMMARY. A chemical and petrographic study of charnockites and pyroxene/hornblende-bearing granulites has been undertaken, and the data used in an interpretation of the petrogenesis of these rocks. Major- and trace-element analyses of fourteen rocks are presented, together with major-element analyses of seven orthopyroxenes, seven clinopyroxenes, two garnets, and three hornblendes. The compositions of the coexisting ferromagnesian minerals are used to obtain estimates of the likely pressure/temperature conditions of equilibration (5–7 kbar and 700±50 °C) of the various mineral assemblages.

Geological setting. The Precambrian rocks of Ceylon are divided into the *Highland Series* (metasediments and charnockites) and the *Vijayan Series* (granites, granitic gneisses, and migmatites)—Cooray (1962). The area studied in the south-east of Sri Lanka straddles the boundary between the Highland Series and the Vijayan Series, the most significant features being the absence of a transitional zone and the presence of acid charnockites in the form of circular domes in the Vijayan Series. This is in contrast to the findings of Cooray (1961), who mapped an area north of the present area of study.

The prominent rock types encountered are: Highland Series: charnockites (acid, intermediate, and basic); undifferentiated metasediments (garnetiferous granulites, garnetiferous biotite gneisses, garnet–sillimanite–biotite gneisses); khondalites (quartz–feldspar–garnet–sillimanite–graphite schists); quartzites; crystalline limestones; calc-gneisses and calc-granulites; amphibole granulites; and amphibolites. These rocks are found closely associated with each other and this association is invariably of an interbanded nature.

The area immediately east of the Highland Series is underlain by rocks grouped under the Vijayan Series (Balendran, 1968): hornblende–biotite gneisses; granites; granitic gneisses and migmatites; acid to intermediate charnockites; calc-granulites

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and calc-gneisses; and amphibolites. The hornblende–biotite gneisses are the main lithological unit in this area and the granites and pink granite gneisses have been demarcated purely on their field relations to the hornblende–biotite gneisses. Acid to intermediate charnockites occur as circular domes and are in a different environment as compared to those of the Highland Series, where the charnockites are mostly associated with metasediments. Charnockites are rare in the Vijayan Series and were regarded by Cooray (1962) as relicts of an earlier metamorphic event.

In a recent examination of a Gondwanaland re-assembly of Peninsular India and part of east Antarctica, Sri Lanka is accommodated, without displacement relative to India, in the Antarctic Ocean re-entrant immediately off Lützow-Holm Bay, which is composed of a charnockitic group and a granitic group (Tatsumi and Kizaki, 1969). The recent geological mapping of the south-eastern coastal belt has revealed a similar lithological association in the Vijayan Series (Balendran, 1968).

Geochronology and metamorphism. The interpretation of existing structural, metamorphic, and isotopic-age data relevant to the question of the geological relationship between the Highland Series and Vijayan Series rocks remains controversial (see, for example, Cooray, 1961, 1962; Katz, 1971; Berger, 1973).

Vijayan Series rocks dated by Crawford and Oliver (1969) define a whole-rock isochron at 1150 ± 60 Myr—the suggested date for the regional scale almandine–amphibolite facies metamorphism and migmatization, which have affected these rocks. Unfortunately the charnockites found as scattered occurrences within the eastern Vijayan have not so far been dated.

Charnockites from the Highland Series have yielded Rb/Sr ages (Crawford and Oliver, 1969) ranging from roughly 1250 to 3000 Myr. They regarded the older ages as relicts of earlier events, suggesting that there have been several episodes of metamorphism affecting these rocks and leading to charnockite development. Alternatively the range of dates could perhaps be interpreted as the result of varying degrees of isotopic re-equilibration of old (at least 3000 Myr) metamorphic rocks during the most recent major metamorphic event (about 1250 Myr). In either case it seems probable that the Highland Series rocks underwent fairly extensive recrystallization and isotopic homogenization under still essentially granulite facies conditions at roughly the same time as the Vijayan Series rocks suffered regional scale almandine–amphibolite facies metamorphism.

Petrography. The rock types studied in detail were charnockites (acid, intermediate, and basic types as defined by Cooray, 1962), pyroxene granulites, and an amphibole granulite.

The charnockites from the Highland Series vary from acid to basic types. Both intermediate and basic charnockites contain coexisting ortho- and clino-pyroxenes, as do the pyroxene granulites although in greater quantities. Biotite is sometimes present and hornblende almost invariably. The hornblende, both in green and brown varieties, usually occurs closely associated with pyroxene. Whilst in many cases it appears to be of late development, in other instances it may well be of earlier origin, as, for example, where rimmed by orthopyroxene in one rock. Garnet is a frequent constituent in the basic charnockites. Plagioclase shows a wide range in composition

(An_{16-60}) and the potash feldspar is commonly a microperthitic orthoclase. The amphibole granulite sample studied has a very high modal content of hornblende and clinopyroxene with an absence of orthopyroxene and garnet.

The acid charnockites from the Vijayan Series are characterized by an abundance of quartz and feldspar with small amounts of highly altered subidiomorphic orthopyroxene, and a total absence of both clinopyroxene and garnet. These charnockites differ from those of the Highland Series in their much higher modal contents of perthitic orthoclase. The perthites are mainly of the replacement type and generally show a myrmekitic intergrowth at plagioclase margins.

TABLE I. *Modal mineralogy of charnockites and pyroxene/amphibole granulites from SE. Sri Lanka*

	Highland Series									Vijayan Series				
	1	2	3	4	5	6	7	8	9	10	11	12	13	14
Quartz	—	—	—	—	14	7	10	28	Maj	Maj	7	20	26	—
K Feldspar	4	1	2	2	2	8	4	13	Maj	Maj	34	59	50	2
Plagioclase	28	9	31	47	38	55	39	26	Maj	Maj	27	15	13	32
Biotite	—	—	—	2	4	10	12	2	Acc	Maj	10	—	1	2
Garnet	10	Tr	15	—	17	—	—	Tr	Min	—	—	—	—	—
Hornblende	15	5	5	16	9	1	14	—	Min	Min	8	3	5	45
Orthopyroxene	18	30	18	14	5	8	6	28	Acc	Acc	2	2	3	—
Clinopyroxene	20	55	27	16	9	9	11	—	—	—	—	—	—	17
Opaques	5	1	2	2	2	2	4	2	Acc	Acc	2	1	2	2
% An in Plag.	46	48	56	40	60	36	36	42	30	12	14	12	12	36

Maj = Major minerals > 10 % Min = Minor minerals 2–10 %
Acc = Accessory minerals <2 % Tr = Trace <0·5 %

1. Basic charnockite, 48/116.
2. Pyroxene granulite, 49/87/1.
3. Pyroxene granulite, J/50/2.
4. Basic charnockite, 49/16/1.
5. Basic charnockite, 48/28/3.
6. Intermediate charnockite, 42/7.
7. Intermediate charnockite, 48/104/1.
8. Acid charnockite, 42/81/1.
9. Acid charnockite, J/49/2.
10. Acid charnockite, J/51/7.
11. Acid charnockite, J/51/2/2.
12. Acid charnockite, J/51/10.
13. Acid charnockite, J/51/5.
14. Amphibole granulite, 48/64.

From Table I it is apparent that the samples analysed in this study fall into the following mineral assemblages: orthopyroxene–clinopyroxene–garnet–hornblende±biotite–plagioclase; clinopyroxene–hornblende–biotite–plagioclase; and orthopyroxene±hornblende±biotite–plagioclase. K feldspar is invariably present and quartz commonly so.

Rock chemistry. The major- and trace-element analyses of the charnockites and their associated rocks are presented in Table II. When plotted on an ACF diagram all the rocks fall within the diopside–anorthite–hypersthene field corresponding to the granulite facies assemblages observed. The low Fe_2O_3/FeO ratio for all samples suggests low partial pressures of oxygen (Goldschmidt, 1954). A general tendency for MgO, CaO, and FeO contents to increase with basicity was noted.

When the chemical changes in the rocks are plotted on variation diagrams based on

Larsen's method, modified by Nockolds and Allen (1953), there is a marked change in the trends of K, Na, and Al curves for the Vijayan Series and Highland Series charnockites and granulites. This trend is also evident from fig. 1 where the basic charnockites plot close to basaltic rocks and the compositional trend from basic to acid members is towards increasing K. The AFM diagram (fig. 2) also illustrates the compositional trend from basic to acid charnockites with the increasing alkali content.

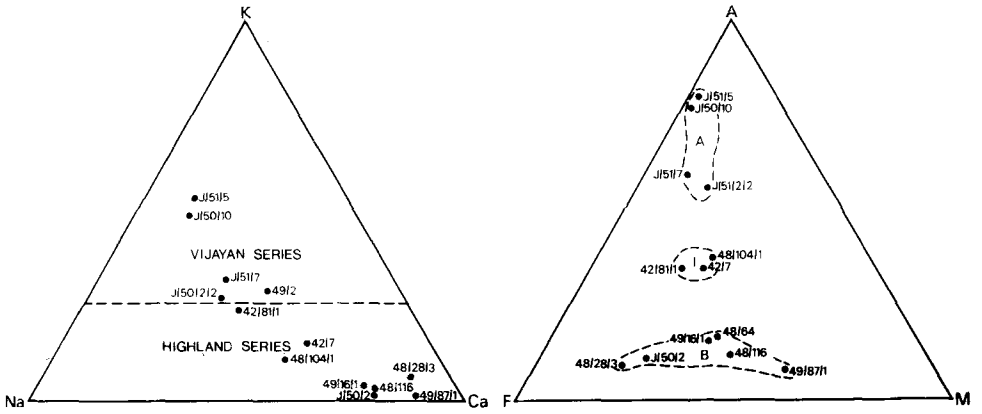
TABLE II. *Chemical analyses of charnockites and pyroxene/amphibole granulites from SE. Sri Lanka*

	Highland Series									Vijayan Series				
	1	2	3	4	5	6	7	8	9	10	11	12	13	14
SiO_2	45·50	48·81	47·77	48·21	50·87	53·90	56·12	66·47	70·62	64·02	64·90	68·59	72·09	48·89
TiO_2	1·96	0·42	2·46	1·77	2·84	1·38	0·89	1·24	0·84	0·69	0·49	0·40	0·41	1·29
Al_2O_3	13·55	13·41	13·06	13·20	12·95	16·19	17·55	12·21	11·66	16·57	14·90	14·49	13·05	13·95
Fe_2O_3	4·08	2·34	0·00	3·95	6·30	3·45	2·70	1·74	1·62	1·06	1·18	1·88	1·03	3·10
FeO	13·07	7·22	16·91	9·79	10·90	6·13	4·97	6·30	4·68	3·81	3·56	2·51	1·91	8·90
MnO	0·20	0·18	0·21	0·22	0·28	0·17	0·14	0·12	0·13	0·10	0·09	0·12	0·03	0·19
MgO	7·30	12·16	5·95	7·58	2·98	3·95	3·77	2·83	1·00	1·29	2·02	0·12	0·27	7·45
CaO	10·92	12·80	10·50	10·58	8·89	7·87	6·79	3·19	3·64	3·53	3·71	1·59	1·37	12·10
Na_2O	2·50	1·42	2·53	2·85	0·89	3·70	4·27	3·17	2·51	4·42	4·74	4·53	3·79	2·32
K_2O	0·40	0·13	0·26	0·50	0·57	1·77	1·14	1·75	2·20	3·30	2·72	5·13	5·38	1·00
P_2O_5	0·30	0·04	0·40	0·19	1·53	0·72	0·28	0·28	0·17	0·24	0·11	0·06	0·08	0·13
SO_3	0·46	0·00	0·40	0·72	0·36	0·07	0·05	0·09	0·10	0·07	0·09	0·03	0·02	0·36
H_2O^+	0·34	0·39	0·11	0·28	0·27	0·33	0·32	0·14	0·13	0·12	0·28	0·18	0·20	0·38
H_2O^-	0·10	0·22	0·11	0·16	0·05	0·17	0·07	0·10	0·09	0·12	0·13	0·09	0·04	0·07
Total	100·68	99·54	100·67	100·00	99·68	99·80	99·06	99·63	99·39	99·34	98·92	99·72	99·67	100·13
Trace elements (in ppm):														
Ba	128	58	91	58	662	1218	625	538	—	—	347	497	407	—
Co	80	64	71	54	49	34	28	32	—	—	11	6	3	—
Cr	175	283	160	166	120	52	95	129	—	—	121	22	28	—
Cu	78	32	117	98	206	142	31	28	—	—	13	9	5	—
Mn	1779	1387	1642	1680	1753	1188	1199	871	—	—	693	717	378	—
Ni	116	64	90	44	19	115	72	63	—	—	24	3	10	—
Rb	5	2	3	7	23	42	18	51	—	—	92	96	176	—
Sr	246	137	130	127	142	651	660	211	—	—	275	111	69	—
V	426	227	378	336	69	232	166	119	—	—	55	—	11	—
Y	40	14	59	38	102	38	28	32	—	—	49	94	99	—
Zn	124	101	131	135	179	95	79	68	—	—	91	73	69	—
Zr	138	19	157	73	526	131	107	223	—	—	214	609	297	—

Both major and trace elements determined by X-ray fluorescence analysis, except for Na_2O, H_2O, and FeO, which were determined by atomic absorption, gravimetric, and Wilson's vanadium-reagent–titration methods, respectively. Analyst: D. E. de S. Jayawardena.

The column numbers equate with those in Table I, for which a sample key is provided.

Fig. 3 shows the variation in content of various trace elements plotted against the major element differentiation index $(\frac{1}{3}Si+K)-(Ca+Mg)$ for the analysed rock suite. It is clear that there is a relative enrichment in Cr, Ni, V, and to a lesser extent Co in the basic charnockites and pyroxene granulites. Zr is low in the basic charnockites but increases in the intermediate group; however, values drop somewhat in the more acid rocks from the Highland Series, although they rise to a peak again in the acid charnockites of the Vijayan Series. Sr increases rapidly with increasing plagioclase content in the basic to intermediate charnockites and then decreases towards the acid end. Highest Ba contents are likewise found in the intermediate charnockites, whilst



FIGS. 1 and 2: FIG. 1 (left). K:Na:Ca variation diagram for the analysed charnockites/granulites from SE. Sri Lanka. FIG. 2 (right). AFM variation diagram illustrating chemical distinctions between the analysed acid charnockites (A), intermediate charnockites (I), basic charnockites and granulites (B).

Rb is at its highest in the acid types. The content of Zn and Cu was found to be generally uniform with a slight increase in the intermediate types.

The variation of major and trace elements for acid to basic charnockites and pyroxene granulites is generally similar to that shown by the Madras charnockite series (Howie, 1955). In general the acid charnockites from the Vijayan Series show a depletion in Ni, Cr, Co, V, and Sr, and are enriched in Zr and Rb as compared to the basic to intermediate charnockites of the Highland Series. There is no significant variation in the trace elements in acid charnockites from the Highland and Vijayan Series.

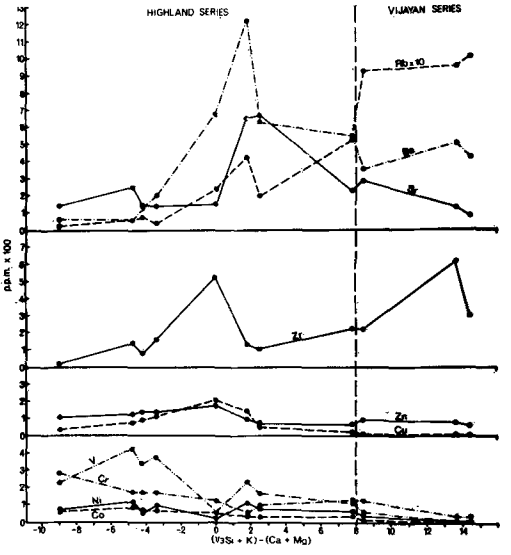


FIG. 3. A plot showing the variation in trace-element content against differentiation index for the analysed Highland Series and Vijayan Series charnockites/granulites.

The determined major and trace elements give a broad indication that the basic charnockites closely resemble basaltic rocks and the variation from the basic to acid members is similar to that shown by a normal igneous differentiation series probably of a calc-alkaline nature. However, the marked increase in Na, K, Rb, and to a lesser extent Al in the Vijayan Series charnockites may reflect some form of alkali metasomatism or granitization, as these rocks are closely associated with granites and migmatites in the field.

Mineral chemistry

Orthopyroxenes. The analytical results for major oxides in seven orthopyroxenes are presented in Table III, together with their structural formulae. The samples were all from the acid to basic charnockites and pyroxene granulites of the Highland Series. The 100 Mg/(Mg+Fe+Mn) ratios calculated vary from 46·0 to 67·9, compared with 35·3–49·0 for orthopyroxenes from the Madras charnockite series (Howie, 1955).

Alumina is generally lower than in the Madras charnockite series and varies from 0·98 wt. % Al_2O_3 for J/50/2 (pyroxene granulite—with garnet) to 1·86 wt. % Al_2O_3 (acid charnockite). However, the orthopyroxene in pyroxene granulite 49/87/1 has an appreciably higher Al_2O_3 content of 4·34 wt. %.

The experimental work of Green and Ringwood (1967) has indicated that the Al_2O_3 content of coexisting pyroxenes may be expected to increase from low- to high-pressure granulite assemblages for rocks of comparable chemistry. However, this is only true for garnet-free assemblages. Once garnet becomes stable in a particular rock, the Al_2O_3 content of the pyroxenes will in fact begin to drop with increasing pressure as further garnet forms (Boyd and England, 1964). Clearly the Al_2O_3 contents of granulite pyroxenes are controlled not only by the *PT* conditions of crystallization, but by chemical control on mineralogy (Howie, 1965).

As regards the pyroxene granulite 49/87/1 the effect of the higher Mg/Fe and Ca/Na ratios in this rock, compared with the other analysed rocks, appears to have been to restrict the content of highly aluminous phases, namely garnet and plagioclase, under the prevailing *PT* conditions of formation, thus resulting in the presence of more aluminous pyroxenes in this rock. Such an explanation is in line with the experimental evidence of Green and Ringwood (1967), which indicated that under granulite facies *PT* conditions garnet appears at lower pressures in rocks with lower Mg/Fe ratios, and plagioclase is stable to higher pressures in rocks with lower Ca/Na ratios.

Clinopyroxenes. The results of chemical analyses for major oxides in seven clinopyroxenes are presented in Table III, together with their structural formulae. In these clinopyroxenes the amount of tetrahedral aluminium is generally greater than octahedral aluminium, the replacement of Si by Al not exceeding 7 %. In the majority of clinopyroxenes Al_2O_3 does not exceed 3 wt. %, but in 49/87/1 the clinopyroxene has an appreciably higher Al_2O_3 content (4·55 wt. %) as did its associated orthopyroxene as discussed above.

The CaO content of the clinopyroxene is fairly constant varying from 20·73 wt. % to 21·06 wt. %, except for 49/87/1, which has a significantly higher CaO content of 22·19 wt. %. Although temperature has a major effect on the extent of solid solution between coexisting pyroxenes (Boyd and Schairer, 1964; Davis and Boyd, 1966), increasing Fe/Mg ratio in the pyroxenes may also be expected to lead to progressive shrinkage of the two-pyroxene field (Lindsley and Munoz, 1969). Thus the higher CaO content of the clinopyroxene in 49/87/1 does not necessarily indicate a lower crystallization temperature than in the other analysed rocks, but may just reflect the significantly lower Fe/Mg ratio in this rock and its constituent minerals.

Two samples of *garnets* were analysed from a pyroxene granulite (J/50/2) and a

TABLE III. *Chemical analyses of pyroxenes*

	Orthopyroxenes							Clinopyroxenes						
	1	2	3	4	5	6	7	1′	2′	3′	4′	5′	6′	7′
SiO_2	49·14	49·15	49·51	49·60	50·50	50·60	50·92	51·50	51·17	50·50	50·54	50·87	50·53	50·49
TiO_2	0·30	0·12	0·12	0·14	0·11	0·13	0·10	0·20	0·23	0·28	0·24	0·21	0·28	0·47
Al_2O_3	0·98	1·86	1·59	1·50	1·27	1·55	4·34	2·14	2·41	2·61	2·09	2·10	2·79	4·55
Fe_2O_3	—	—	—	—	—	—	—	1·54	1·87	3·57	4·02	2·63	3·06	2·28
FeO	33·40*	31·93*	31·34*	30·43*	25·30*	27·63*	19·69*	12·06	10·40	10·46	8·61	8·18	8·21	5·00
MnO	0·33	0·53	0·46	0·91	1·03	0·59	0·34	0·17	0·42	0·26	0·39	0·50	0·24	0·17
MgO	13·93	16·14	15·21	16·30	18·38	18·28	23·79	10·94	12·03	11·13	11·87	12·84	12·36	14·16
CaO	0·92	0·42	0·96	0·98	1·05	0·71	1·07	20·74	20·89	20·73	20·89	20·73	21·06	22·19
Na_2O	0·02	0·01	0·03	0·01	0·04	0·01	0·02	0·42	0·41	0·46	0·46	0·54	0·47	0·54
K_2O	0·01	0·01	0·01	0·03	0·04	0·01	0·01	0·04	0·05	0·02	0·03	0·07	0·03	0·01
P_2O_5	0·01	0·00	0·00	0·00	0·01	0·01	0·00	0·04	0·02	0·04	0·03	0·03	0·04	0·02
SO_3	0·00	0·00	0·01	0·06	0·01	0·00	0·02	0·03	0·09	0·08	0·07	0·09	0·03	0·04
Total	99·04	100·17	99·24	99·96	97·74	99·52	100·30	99·82	99·99	100·14	99·24	98·79	99·10	99·92
Cation proportions on the basis of 6 oxygens:														
Si	1·960	1·922	1·950	1·937	1·967	1·949	1·874	1·956	1·933	1·914	1·922	1·933	1·914	1·870
Al^{iv}	0·040	0·078	0·050	0·063	0·033	0·051	0·126	0·044	0·067	0·086	0·078	0·067	0·086	0·130
Al^{vi}	0·006	0·008	0·074	0·006	0·025	0·019	0·062	0·052	0·040	0·031	0·016	0·027	0·039	0·069
Ti	0·009	0·004	0·004	0·004	0·003	0·004	0·003	0·006	0·007	0·008	0·007	0·006	0·008	0·013
Fe^{3+}	—	—	—	—	—	—	—	0·044	0·053	0·102	0·115	0·075	0·087	0·064
Mg	0·828	0·941	0·893	0·949	1·067	1·049	1·305	0·619	0·677	0·629	0·673	0·727	0·698	0·781
Fe^{2+}	1·114	1·004	1·032	0·994	0·894	0·890	0·606	0·383	0·329	0·332	0·274	0·260	0·260	0·155
Mn	0·011	0·018	0·015	0·030	0·034	0·019	0·011	0·005	0·013	0·008	0·013	0·016	0·008	0·005
Ca	0·039	0·018	0·041	0·041	0·044	0·029	0·042	0·844	0·845	0·842	0·851	0·844	0·855	0·880
Na	0·002	0·001	0·002	0·001	0·003	0·001	0·001	0·031	0·030	0·034	0·034	0·040	0·035	0·039
K	0·001	0·000	0·001	0·001	0·002	0·000	0·000	0·002	0·002	0·001	0·001	0·003	0·001	0·000
Σ (Al^{vi} to Mn)	1·968	2·015	2·018	1·983	1·953	1·981	1·987	1·109	1·119	1·110	1·098	1·111	1·100	1·087
Σ (Ca, Na, K)	0·042	0·019	0·044	0·043	0·049	0·030	0·043	0·877	0·877	0·877	0·886	0·887	0·891	0·919
Mg	41·80	46·98	45·42	47·83	55·14	53·30	66·82	32·82	35·65	33·18	35·37	38·09	36·42	41·92
Fe	56·23	52·12	52·49	50·10	42·58	45·22	31·03	22·43	19·85	22·42	19·92	17·71	17·92	11·32
Ca	1·97	0·90	2·09	2·07	2·27	1·47	2·15	44·75	44·50	44·40	44·70	44·21	45·19	46·76

* Total Fe value (XRF) quoted as FeO in orthopyroxenes. FeO determined by Wilson's procedure in clinopyroxenes. Na_2O by atomic absorption; all other elements by X-ray fluorescence. Analyst: D. E. de S. Jayawardena.

All pyroxene separates from Highland Series rocks:

1 and 1′ from pyroxene granulite J/50/2.
2 from acid charnockite 42/81/1.
2′ from amphibole granulite 48/64.
3 and 3′ from basic charnockite 48/116.
4 and 4′ from intermediate charnockite 42/7.
5 and 5′ from intermediate charnockite 48/104/1.
6 and 6′ from basic charnockite 49/16/1.
7 and 7′ from pyroxene granulite 49/87/1.

basic charnockite (48/28/3) and the analyses are presented in Table IV. These garnets were found to be richer in andradite, grossular, and spessartine, and poorer in almandine when compared to garnets from the gneisses (Jayawardena, 1973). The low MnO content, (FeO+MnO)/MgO ratio, and the almandine content of the garnets are in agreement with those from similar metamorphic terraines (Miyashiro, 1953; Sturt,

TABLE IV. *Chemical analyses of garnets and partial analyses of hornblendes*

	Garnets		Hornblendes				Structural formulae				
	1	2	3	4	5		1′	2′	3′	4′	5′
SiO_2	37·94	37·99	41·91	41·86	40·38	Si	3·005	3·036	6·264	6·247	6·088
TiO_2	0·10	0·14	2·24	2·18	2·60	Al^{iv}	—	—	1·736	1·753	1·912
Al_2O_3	20·28	20·30	11·63	11·13	12·38	Al^{vi}	1·893	1·912	0·312	0·204	0·288
Fe_2O_3	2·84	0·39	4·72	4·67	4·26	Ti	0·006	0·008	0·252	0·245	0·295
FeO	24·87	28·20	11·81	13·09	14·79	Fe^{3+}	0·169	0·023	0·531	0·524	0·483
MnO	0·95	0·82	0·17	0·23	0·91	Fe^{2+}	1·648	1·885	1·476	1·634	1·865
MgO	4·51	4·01	10·75	11·24	9·05	Mn	0·064	0·478	0·022	0·029	0·116
CaO	7·51	6·85	11·57	11·40	11·11	Mg	0·533	0·056	2·395	2·500	2·034
Na_2O	0·01	0·04	1·45	1·12	2·06	Ca	0·637	0·586	1·853	1·823	1·795
K_2O	0·01	0·02	1·52	1·53	1·01	Na	0·002	0·006	0·420	0·324	0·602
						K	0·001	0·002	0·290	0·291	0·194
Total	99·02	98·76	97·77	98·45	98·55	O	12·000	12·000	23·000	23·000	23·000
					% Almandine		57·2	61·4	*X* 2·563	2·438	2·591
					% Andradite		8·8	1·2	*Y* 4·986	5·136	5·081
					% Grossular		13·3	19·0	*Z* 8·000	8·000	8·000
					% Pyrope		18·5	16·4			
					% Spessartine		2·2	1·9			

All elements determined by X-ray fluorescence analysis except Na_2O (atomic absorption) and FeO (Wilson's vanadium-reagent method). Analyst: D. E. de S. Jayawardena.

All mineral separates from Highland Series rocks:

1 and 1′ garnet from basic charnockite 48/28/3.
2 and 2′ garnet from pyroxene granulite J/50/2.
3 and 3′ hornblende from basic charnockite 49/16/1.
4 and 4′ hornblende from amphibole granulite 48/64.
5 and 5′ hornblende from basic charnockite 48/116.

1962; Engel and Engel, 1960). It is, however, apparent that the host-rock composition plays an important role in controlling the chemical composition of garnets and it is difficult to separate the variation due to host rock from that due to grade of metamorphism.

Three samples of *hornblendes* from 48/64 (amphibole granulite) and 48/116 and 49/16/1 (basic charnockites) were analysed (Table IV). Their compositions lie between the fields of common hornblende and pargasite. When their compositions are calculated as pargasite–ferropargasite and magnesiohastingsite–hastingsite end-members (Gilbert, 1966), they plot diagonally across the end-members pargasite and hastingsite and exhibit less scatter than those from Madras (Howie, 1955). The compositions of the hornblendes studied are similar to those from Kondapalli, India (Fig. 1, Leelanandam, 1970).

In the hornblendes of the present study the number of Ca atoms per formula unit varies from 1·87 to 1·93 and falls within the hornblende field (Deer *et al.*, 1961). The replacement of Mg by Fe^{2+} shows a wide variation when expressed as 100 $Mg/(Mg+Fe^{2+}+Fe^{3+}+Mn)$ and Fe^{3+}/Fe^{2+} is less than 1. The relatively high TiO_2 contents and Al^{vi} values in hornblendes from basic charnockites and amphibole granulites as compared to hornblendes from gneisses and amphibolites suggest somewhat higher temperatures and pressures of crystallization for the Highland Series (Jayawardena, 1973). However, it is difficult to distinguish between the compositional control imposed by the whole rock chemistry and that due to the prevailing temperatures and pressures of crystallisation.

Petrogenesis: Temperature estimates

Coexisting pyroxenes. Six pairs of coexisting pyroxenes have been analysed from Highland Series charnockites/granulites. In all cases, the Ca-rich pyroxenes have lower Fe^{2+}/Mg^{2+} ratios than the associated Ca-poor pyroxenes. Hence pyroxene tie-lines on the En–Wo–Fs diagram cut the En–Wo side-line when extended, as to be expected for assumed equilibrium assemblages (Howie, 1965).

The Mg^{2+}–Fe^{2+} distribution coefficient K_D (Kretz, 1961) or K_p (Bartholomé, 1962) between the coexisting pyroxenes can be used to estimate the likely temperature of equilibration of these mineral assemblages. The range of K_D values (Table V) is rather large, particularly with the analytically determined Fe^{2+} values—no doubt at least in part reflecting the difficulty in obtaining accurate Fe^{2+}/Fe^{3+} ratios with standard geochemical techniques as employed here. A comparison with temperature estimates and corresponding K_D values for coexisting pyroxenes in rocks for Madras (Howie, 1955, 1965), Colton area (New York; Engel *et al.*, 1964), Saltora (India; Sen and Rege, 1966), and Amaravathi (Andhra Predesh; Ramaswamy and Murty, 1973), suggest equilibration temperatures in the range 700 ± 100 °C for the mineral assemblages from south-east Sri Lanka.

Manna and Sen (1974) have indicated that garnetiferous basic granulites have lower K_D values and thus lower equilibration temperatures as compared to associated garnet-free basic granulites in the Saltora district of West Bengal. However, the values in Table V do not show any systematic differences in K_D between the garnetiferous and garnet-free granulites studied here. It is, however, apparent that higher whole-rock Fe/Mg ratios seem to have favoured the formation of garnet in certain of these basic charnockites under the prevailing *PT* conditions. This is in line with the experimental evidence of Green and Ringwood (1967). However, it has to be pointed out that the samples in this study were collected over a wide area, unlike the rock suite studied by Manna and Sen (1974), hence regional variation in the *PT* conditions of metamorphism are more likely in this instance.

The $Ca/(Mg+Fe^{2+})$ ratio in the clinopyroxenes can also be used to give an estimate of the temperature of crystallization by comparison with solid-solution limits for the coexisting pyroxene pairs in the synthetic $MgSiO_3$–$CaMgSi_2O_6$ system (O'Hara and Mercy, 1963; Boyd and Schairer, 1964). It has been shown experimentally (Davis and Boyd, 1966) that pressure has little effect on the position of the diopside solvus. The

solvus limits extrapolated on to the $MgSiO_3$–$CaMgSi_2O_6$ join for the curves representing the two-pyroxene field of the present study are approximately Ca_1Mg_{99} and $Ca_{48}Mg_{52}$ (Jayawardena, 1973) and give an approximate temperature of equilibration of 850±150 °C (Boyd and Schairer, 1964). Unfortunately this geothermometer is rather insensitive at such temperatures, as the solvus boundaries become almost parallel to the temperature axis at low temperatures. The semi-empirical method of Wood and Banno (1973) for calculating the equilibrium temperatures from the coexisting pyroxene data gives closely similar values (858±11 °C as the mean temperature and standard deviation for the six pyroxene pairs).

TABLE V. Ca/(Ca+Mg) *ratios and* K_D *values for coexisting pyroxenes*

Sample Number	CPX			OPX			KD	
	Ca	Mg	$\frac{Ca}{Ca+Mg}$	Ca	Mg	$\frac{Ca}{Ca+Mg}$	*Calc. Fe^{2+}	†Det. Fe^{2+}
J/50/2	44·75	32·82	0·577	1·97	41·80	0·045	0·514	0·519
48/116	44·40	33·18	0·572	2·09	45·42	0·044	0·551	0·521
42/7	44·70	35·37	0·558	2·07	47·83	0·041	0·505	0·450
48/104/1	44·21	38·09	0·537	2·27	55·14	0·040	0·550	0·505
46/16/1	45·91	36·42	0·558	1·47	53·30	0·027	0·528	0·476
49/87/1	46·76	41·92	0·527	2·15	66·82	0·031	0·500	0·524

* These K_D values are based on Fe^{2+} values obtained following calculation of the Fe^{2+}/Fe^{3+} ratios in these pyroxenes from the structural formulae assuming stoichiometry, by a similar procedure to that used by Mysen and Heier (1972).

† These K_D values are based on the determined Fe^{2+} values.

Coexisting garnet and clinopyroxene. Only one coexisting garnet–clinopyroxene pair has been analysed in this study, but the partitioning of Fe^{2+} and Mg between these mineral phases can also be used to give an indication of the equilibration temperature. Banno (1970) used the derived distribution coefficient K' to obtain a rough estimate of temperature for the purpose of a genetic classification of eclogites and according to him the pressure effect on K' is negligible.

The distribution coefficient K' calculated for sample J/50/2 is 6·37 and using the compiled analyses and temperature estimates for calibration of the $K' = f(T)$ geothermometer of Mysen and Heier (1972) a value of 670 °C can be deduced. Råheim and Green (1974) have demonstrated through experimental work that K' is more highly dependent on pressure than originally thought by Banno (1970), and their derived expression for relating K' to equilibration temperature gives a value of 656 °C if a pressure of 7 kb is assumed as a first approximation.

As errors in K', and likewise K_D, can be introduced by inaccurate Fe^{2+}/Fe^{3+} ratio determinations, it is also worth considering the distribution coefficient K'' for Mn and Mg between the garnet and clinopyroxene. The calculated K'' for J/50/2 is 15·33 and from fig. 4 (Carswell, 1974) an approximate temperature of 675 °C can be estimated. The temperature estimates derived from the Fe^{2+}/Mg and Mn/Mg partition coefficients

are thus in close agreement, although it is realistic to put error limits of at least ±50 °C on these values.

It is apparent that there is a discrepancy of close to 200 °C between the values derived from consideration of the Ca distribution between the coexisting pyroxenes and from the Fe^{2+}–Mg–Mn distribution between coexisting garnet and clinopyroxene. It is considered that the higher estimates based on the former method are more likely to be in serious error and hence that a value of 700±50 °C is probably most appropriate for the final equilibration of these assemblages. As the various estimates are based on analyses of bulk mineral separates, it may be that the range of temperature values reflect the temperature range from that of initial formation down to final equilibration of the charnockite/granulite terrain during subsequent cooling/unloading.

Pressure estimates

Green and Ringwood (1967) carried out an experimental investigation of the gabbro to eclogite transformation, resulting in a subdivision of basic granulite assemblages into low-, intermediate-, and high-pressure types.

The SiO_2 undersaturated pyroxene granulites and charnockites studied are characterized by orthopyroxene+clinopyroxene+garnet+plagioclase and orthopyroxene+clinopyroxene+plagioclase assemblages. The Lewisian metamorphic rocks of Scourie, Sutherland, described by O'Hara (1961) similarly contain the assemblage orthopyroxene+clinopyroxene+garnet+plagioclase in undersaturated basic rocks. De Waard (1964) described an area in the Adirondacks where he drew attention to the intermingling of orthopyroxene+clinopyroxene+plagioclase+quartz+hornblende and clinopyroxene+garnet+plagioclase+quartz+hornblende assemblages. This intermingling was explained by De Waard (op. cit.) as due to the presence of mineral assemblages from both sides of the reaction orthopyroxene+plagioclase = clinopyroxene+garnet+quartz. This reaction is now recognized as taking place at the boundary between the high-pressure and intermediate-pressure granulite fields. The granulites and charnockites of Madras (Howie, 1955) have a stable mineral assemblage of orthopyroxene+clinopyroxene+plagioclase+quartz+hornblende and this is true for granulite facies terrains in Uganda (Groves, 1935), Varberg (Sweden; Quensel, 1954), and Broken Hill (New South Wales; Binns, 1964).

The assemblage orthopyroxene±clinopyroxene±garnet+plagioclase+quartz± hornblende±biotite in oversaturated rocks of the Highland Series of south-east Sri Lanka therefore appears to have been formed under the *PT* conditions of the intermediate-pressure granulite field (Green and Ringwood, 1967). This is further borne out by the presence of coexisting sillimanite+K-feldspar and absence of muscovite and kyanite in the metasedimentary gneisses that are closely intermixed with the charnockites of the Highland Series.

The charnockites of the Vijayan Series are characterized by the assemblage orthopyroxene+plagioclase+quartz±hornblende±biotite and can therefore also be grouped under the intermediate-pressure granulite field.

The most likely *PT* field of equilibration of the charnockites and pyroxene granulites from both the Vijayan Series and Highland Series is shown on fig. 4. With the favoured

temperature estimates of 700±50 °C, best load-pressure estimates would be in the range 5 to 7·5 kbar. With metamorphic temperatures of that order, the general lack of evidence of crustal partial melting in the Highland Series rocks, the absence of muscovite, and the scarcity of hornblende compared with pyroxenes, together indicate that in general the water pressure must have been significantly less than the total pressure during the granulite facies metamorphism.

Conclusion

From the rock chemistry it is difficult to decide whether the marked increase in Na, K, Rb, and to a lesser extent Al in the Vijayan Series charnockites reflects the typical compositions of the more acid members of a calc-alkaline type differentiation series or whether it is due to alkali metasomatism or granitization during metamorphism. However, the field association of these rocks with granites and migmatites would appear to favour the latter interpretation.

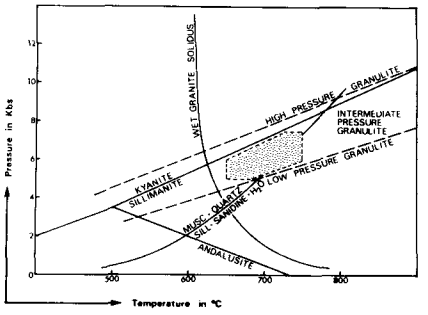


FIG. 4. The stippled field indicates the estimated P/T conditions of equilibration of the SE. Sri Lanka charnockites/granulites in relation to the extrapolated P/T boundaries for the intermediate-pressure granulite field (Green and Ringwood, 1967), the likely stability field of the Al_2SiO_5 polymorphs (Newton, 1966; Fyfe and Turner, 1966; Holdaway, 1971), the experimentally determined reaction curve muscovite+quartz = sillimanite+sanidine+water (Evans, 1965), and the minimum melting curve for water-saturated 'granite' (Bowen and Tuttle, 1958).

Consideration of the partitioning of various elements between coexisting pyroxenes and coexisting clinopyroxene and garnet indicates equilibration temperatures in the range 700±50 °C. Amongst the suite of samples studied there was no evidence that garnetiferous granulites have consistently lower equilibrium temperatures than garnet-free granulites as indicated by Manna and Sen (1974).

The charnockites and pyroxene granulites studied from both the Highland Series and Vijayan Series have mineral assemblages that fall into the intermediate-pressure granulite field of Green and Ringwood (1967), suggesting formation at load pressures in the range 5 to 7·5 kbar, corresponding to crustal depths of roughly 17 to 25 kilometres. Water pressure is thought likely to have been in general significantly less than the load pressure during the granulite facies metamorphism.

On the basis of the mineral data presented and the available age dates, we tentatively suggest that the charnockites of both the Highland Series and Vijayan Series may have equilibrated at broadly similar P_{LOAD}/T conditions at roughly the same time (1150–1250 Myr) as the development of the associated Vijayan Series gneisses, migmatites, and granites. The development of the latter rock types on a regional scale we attribute to the prevalence of much higher P_{H_2O} conditions during this metamorphic event in the Vijayan Series rocks than in the Highland Series rocks. There is, however, a need for further studies, including age dates, on the associated charnock-

ites, gneisses, and granites in the Eastern Vijayan Series to help resolve the question of the genetic relationship between these rocks, and between the Highland and Vijayan Series as a whole.

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