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The Geochemical Distribution and Source of Copper in the Metalliferous Mining Region of Southwest England

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In order to test a hypothesis on the origin of copper in the Cornwall district, 270 samples of mafic rocks, 88 samples of sedimentary rocks and 78 samples of granitic rocks have been analysed for their copper contents. Among the mafic rocks, the intrusive microgabbro has a higher mean copper content (59 ± 32 PPM) than the spilitic pillow lavas (43 ± 27 PPM). The abundance of copper in the sedimentary rocks appears to be related to their lithological characters. The average copper content of the grey slates with carbonaceous material (47 ± 16 PPM) is higher than that for the noncarbonaceous grey slates (29 ± 20 PPM) The distribution of copper in the granitic rocks shows a marked relationship with the degree of mineralisation in the region. The average copper content of the granitic rocks from the intensely mineralized region is 73 ± 18 PPM while the granitic rocks from the less mineralized region contain an average of 21 ± 9 PPM copper. Further, it appears that there has been a general enrichment of copper from early granitic stage to the late pegmatites. These geochemical features can be of potential use in selecting intrusive bodies for detailed exploration. The high copper content in the country rock and in the mineralized intrusion plus a likelihood of assimilation of crust in forming granitic magmas suggest that the copper deposited as hydrothermal lodes may have been derived from the country rocks of the region.

INTRODUCTION

As part of a study aimed at the understanding of the genesis of copper ores in Southwest England, a number of rock types from localities in south Devon and Cornwall were analyzed for the purpose of studying distribution of copper within the rocks of the region. The theory that the source of copper forming hydrothermal deposits lies in the granitic rock of the region is brought into question by the fact that the sedimentary and mafic rocks show copper distribution comparable with the average values of the granites, and also because copper being a chalcophile element is not expected in such large amounts in granites to give rise to hundreds of thousands of tons of copper as ores. While mineralogical and chemical composition of the igneous rocks have been investigated widely in the past, trace metal geochemistry has not been studied in detail. A

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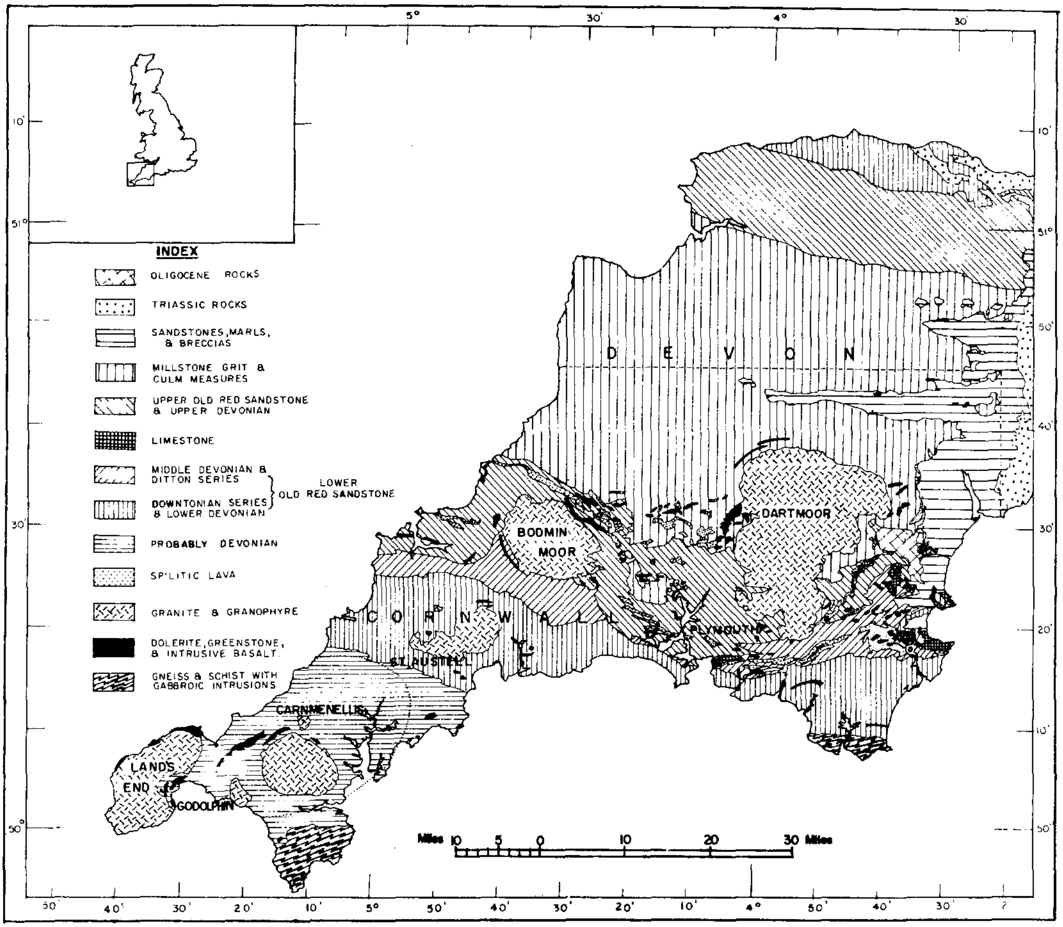

Fig. 1. Map showing the location and general geology of the area (after Geological Survey of England 1/4" map). Richest copper producing region is enclosed by dotted line

few determinations of copper in the region have been made in biogeochemical material (Millman, 1957), and more recently distribution of certain trace elements in minerals from the granites of Southwest England has been described (Bradshaw, 1967). However, a more comprehensive study was necessary before further work on ore genesis could be done. This paper presents the results of more than 430 rock analyses for copper followed by a short discussion on the source of copper for copper mineralization in Southwest England.

Geological Setting

The peninsula of Cornwall and Devon (Southwest England) is composed mainly of a monotonous sequence of greenish grey slates with subordinate sandstones ranging in age from Ordovician to Permian with the occasional development of limestones in the Devonian (Fig. 1) These rocks are believed to have been deposited in a broad geosyncline separated into basins by low ridges (Hosking, 1962) The area is well known for its protracted igneous activity giving rise to the extensive development of intrusive mafic rocks and lava flows varying in age from Ordovician to Permian. The intrusive rocks include microgabbro, proterobase[1], quartz microgabbro and picrite while the extrusive rocks are represented by lavas and associated tuffs. The basaltic lava flows of the Devonian and Carboniferous ages are spilitic while those of Permian age have been extruded under subaerial condition (Tidmarsh 1932, Phillips 1878, Dewy and Flett 1911). In the geological history of the Peninsula, the close of the Carboniferous period is marked by general uplift and a granitic intrusion of batholithic dimension which is now exposed in the form of isolated bosses such as Dartmoor, Bodmin Moor, St. Austell, Carnmenellis and Land's End (Fig. 1). The intrusion of the granitic mass is believed to have taken place in two successive phases (Brammal and Harwood 1932, Gosh 1927, Reid and Flett 1907) giving rise to the older very coarse, strongly porphyritic granite and the younger fine textured granite. Besides the two types of granites, aplite, pegmatite and felsite are found as dykes and veins. The country rocks including the mafic rocks lying close to the granitic bosses have suffered varying degrees of contact metamorphism. The geology of the area and petrology of the rocks have been described in detail by various authors (Hill and McAlister 1906, Reid et al. 1911, Ussher 1902, 1912, 1913, Middleton 1960, Tidmarsh 1932 and Reid and Flett 1907).

The intrusion of the granites has led to the hydrothermal copper mineralization. Copper mining was important in Cornwall and Devon mainly between 1800 and 1890; at present tin is of greater importance. The richest areas of copper mineralization were those around the Land's End and Carnmenellis granitic masses (Fig. 1). The total output of metallic copper from the Cornish mines over the period of 1815 to 1905 was about 726,970 tons while the production from the mines of Devonshire amounted to 71,970 tons for the period 1821 to 1913 (McAlister 1907, 1921), the output from the Cornish mines being 10 times greater. Of the numerous copper minerals found, chalcocite formed by secondary enrichment was by far the most important. Ores are found as zoned lodes with tin at the bottom followed by a mixed copper-tin zone to a rich copper zone through a copper-lead-zinc zone to a lead-zinc zone and ending into iron-lead-zinc and iron zones at the top. The zoned lodes commonly located in the marginal parts of the granitic bodies passed into the surrounding country rocks (Davison 1921, Dines 1956). It is generally agreed that the mineralization is of hydrothermal origin related to Permo-Carboniferous granitic intrusion, though the origin of the metal is open to question.

EXPERIMENTAL WORK

Sampling

Samples totalling 270 mafic igneous rocks, 88 sedimentary rocks and 78 granitic rocks were collected from different localities in the area. An attempt was made to collect from varied exposures so that not only those rock types quarried for economic purposes would be selected but in case of the granites this was not possible so some bias may be involved. Shales or slates and a few limestones were collected from sedimentary sequences, mostly from coastal exposures. During sampling care was taken to avoid any apparent sign of mineralization and weathering so that only fresh rock samples unaffected by mineralization were obtained. This may have introduced certain amount of bias but it was necessary in view of the nature of the intended study.

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[1] The term used in the Geological Survey of England Memoir No. 335-336 (Flett and McAlister, 1910) and described as subalkalic group of rock containing primary brown hornblende and biotite with traces of olivine (SiO_2 42.86 %)

Analytical Method

For chemical analysis, unweathered portions of each rock sample were crushed in a steel mortar to less than 8 mm in size and then ground in an electric mill down to -100 mesh powder of which a 0.25 gram split was used. Silica sand was run after each sample so that contamination of the successive samples by the preceeding ones was avoided.

The sample was taken in a platinum crucible and digested in a mixture of 2 ml of 1:1 H_2SO_4, .5 ml of concentrated HNO_3, 3 ml of HF and a few ml of H_2O for several hours at low temperature followed by evaporation; the sample being reheated after adding a few ml of H_2O and 1:1 H_2SO_4 and finally taken up as a chloride solution as described in detail by Sandell (1959, p. 460). Copper was determined in monovalent state by the colorimetric method described by Sandell using a solution of 2-2 biquinoline in amyl alcohol, the solution finally obtained from the extraction process being measured by spectrophotometer at 545 mμ. The copper concentration was derived from a calibration curve previously determined. Blank experiment performed as a check showed a mean value of 5 PPM copper (maximum 6.8 PPM) present in the reagents used, so all analytical results were reduced by this value.

Precision of the experiments was tested on eleven samples (with varying copper content) by performing each determination five times, giving a mean standard deviation ± 4 PPM due to operator and instrumental error. Accuracy checks were made on the same samples by the addition of a measured 80 PPM copper followed by three determinations. An average of 82 PPM more copper was found instead of 80 PPM copper added with a standard deviation of ± 6.40 PPM; the recovery was judged to be close enough considering the error involved in the preparation of solution and accurate measurement of 80 PPM copper.

RESULTS AND DISCUSSION

A number of samples representing mafic, sedimentary and granitic rocks have been analysed for their copper contents. The distribution of copper in these rocks has been subjected to simple statistical treatment for comparative study. In doing so the acceptable level of significance for the present work has been fixed at 5 %. In cases where the distribution was found to be approximately lognormal, statistical tests were performed on transformed values.

Mafic Rocks

A total of 270 samples of mafic rocks of different types and ages have been analysed for their copper contents; the results are summarized in Table I. Out of the total number (180) of samples of unmetamorphosed mafic rocks only 6 yielded copper content greater than 150 PPM and are assumed to represent a mineralized population as discussed later. These values are 192 PPM (Ordovician), 176 PPM (Devonian), 202 and 254 PPM (Carboniferous) and 175 and 181 PPM (Permian) and have been excluded from the calculation of mean and standard deviation for the various groups of rocks for the reason stated above. Fifty percent of the results show less than 50 PPM copper and about 8 % reveal copper content between 100 and 150 PPM while about 3 % represent copper content greater than 150 PPM. Similarly in case of the metamorphosed rocks 55.5 % of the results yielded less than 50 PPM copper, 8.6 % gave copper content ranging between 100 and 130 PPM while only 2.6 % contain greater than 130 PPM.

Frequency Distribution

Since a frequency histogram of copper in the unmetamorphosed mafic rocks (180 results) showed strong positive skewness, the cumulative frequency distribution was plotted on a log-probability paper which revealed sharp inflections suggesting mixing of popula-

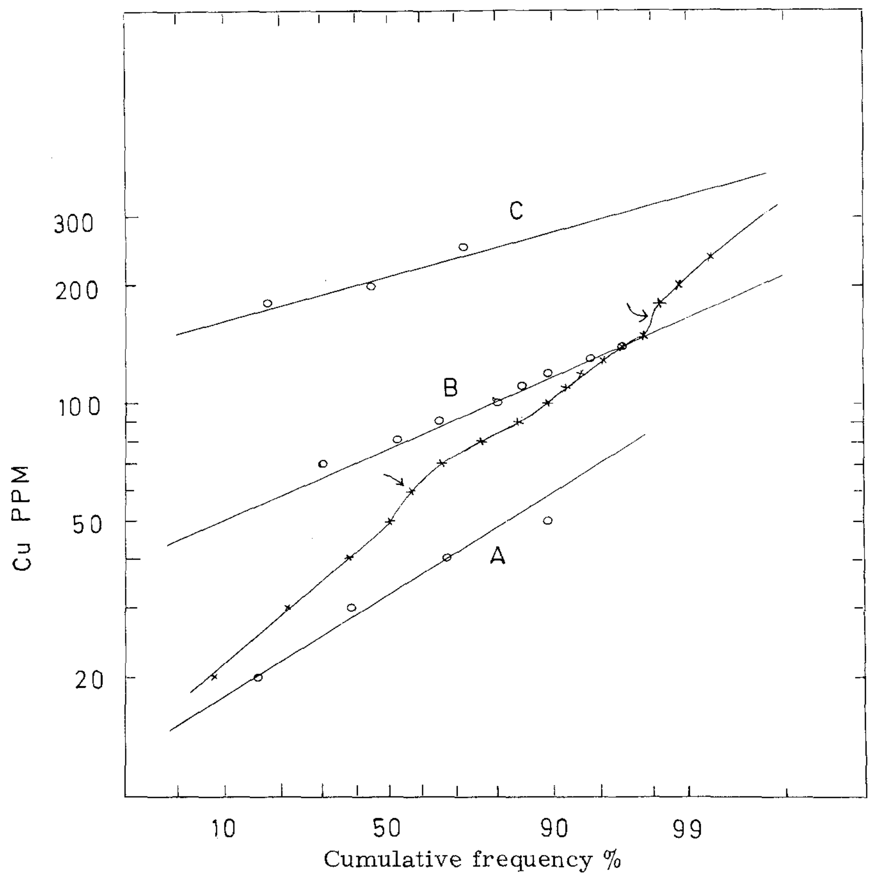

Fig. 2. Cumulative frequency plot and separated populations for the unmetamorphosed mafic rocks. The curve passing through the crosses (based on 180 results) represents original distribution and reveals two inflection points at approximately 56.70 and 98.00 cumulative percentiles indicated by arrow heads. The inflections suggest that the curve results from a combination of three lognormal populations approximated by the three linear subparallel segments of the curve. A, B and C are the three partitioned populations (in the ratio 56.70/41.30/2.00) estimated by the lines passing through the calculated points (open circles)

tions. Therefore, using a method described by Sinclair (1974), the cumulative plot was partitioned into three separate populations (Fig. 2). Population A represents about 56.67 of the entire data and has a mean copper content of 33 PPM while population B constitutes 41.53 % of the data with a mean of 78 PPM. The remaining 2% perhaps represent mineralized population C having a mean copper content of 210 PPM but is not properly represented because of the sampling bias mentioned earlier. Population A with its mean copper content of 33 PPM is best represented by mainly the microgabbro and pillow lavas of unclassified Devonian and upper Devonian ages respectively, whose mean copper contents are 38 PPM and 36 PPM being very close to the mean value obtained for the separated population A. Most of the results for the microgabbro of Ordovician, upper, middle and lower Devonian as well as of Carboniferous periods seem to account for population B. The mean copper content of the calculated popula-

Table 1. Summary of the abundance of copper in the mafic rocks. (Figures in parenthesis indicate parameters based on transformed values)

Rocks	Results	Range PPM	Mean PPM	Standard deviation	Coefficient of variation(%)
Ordovician unmetamorphosed microgabbro	9	17 - 127 (1.23 - 2.10)	76 (1.81)	35 (0.32)	46 (18)
Unclassified Devonian unmetamorphosed microgabbro	16	17 - 58 (1.23 - 1.76)	38 (1.54)	18 (0.21)	47 (14)
Unclassified Devonian metamorphosed microgabbro	30	10 - 113 (1.00 - 2.05)	36 (1.43)	32 (0.33)	89 (23)
Lower Devonian unmetamorphosed microgabbro	16	14 - 120 (1.15 - 2.08)	60 (1.70)	34 (0.33)	57 (19)
Middle Devonian unmetamorphosed microgabbro	10	21 - 98 (1.32 - 1.99)	61 (1.74)	26 (0.24)	43 (14)
Middle Devonian unmetamorphosed quartz microgabbro	4	27 - 51 (1.43 - 1.71)	41 (1.60)	10 (0.13)	24 (8)
Upper Devonian unmetamorphosed microgabbro	54	12 - 148 (1.08 - 2.17)	63 (1.75)	29 (0.23)	46 (13)
Upper Devonian unmetamorphosed proterobase	6	19 - 68 (1.28 - 1.83)	50 (1.66)	20 (0.23)	40 (14)
Upper Devonian unmetamorphosed picrite	5	27 - 45 (1.43 - 1.65)	38 (1.57)	7 (0.09)	18
Upper Devonian metamorphosed microgabbro	15	19 - 121 (1.28 - 2.08)	60 (1.69)	30 (0.26)	50 (15)
Upper Devonian unmetamorphosed pillow lava	19	15 - 79 (1.18 - 1.90)	35 (1.60)	17 (0.23)	49 (14)
Upper Devonian metamorphosed pillow lava	10	40 - 101 (1.60 - 2.00)	64 (1.80)	20 (0.13)	31 (7)
Carboniferous unmetamorphosed microgabbro	29	15 - 144 (1.18 - 2.16)	60 (1.69)	34 (0.29)	57 (17)
Carboniferous metamorphosed microgabbro	24	4 - 116 (0.60 - 2.06)	50 (1.51)	35 (0.51)	70 (34)
Carboniferous unmetamorphosed pillow lava	8	19 - 104 (1.28 - 2.02)	65 (1.73)	34 (0.35)	52 (20)
Permian unmetamorphosed basalt	7	17 - 91 (1.23 - 1.96)	48 (1.62)	27 (0.28)	56 (17)

Table 2. Distribution of copper in various types of unmetamorphosed and metamorphosed mafic rocks. (Figures in parenthesis indicate parameters based on transformed values)

Rocks	Results	Range PPM	Mean PPM	Standard deviation	Coefficient of variation (%)
Unmetamorphosed microgabbro (all ages)	134	10 - 148 (1.00 - 2.17)	59 (1.71)	32 (0.26)	- -
Unmetamorphosed proterobase	6	19 - 68 (1.28 - 1.83)	50 (1.66)	20 (0.23)	- -
Unmetamorphosed quartz microgabbro	4	27 - 51 (1.43 - 1.71)	41 (1.60)	10 (0.13)	- -
Unmetamorphosed picrite	5	27 - 45 (1.43 - 1.65)	38 (1.57)	7 (0.09)	- -
Unmetamorphosed spilitic pillow lava (upper Devonian and Carboniferous)	27	15 - 104 (1.18 - 2.02)	43 (1.57)	27 (0.28)	- -
Unmetamorphosed basalt (Permian)	7	17 - 91 (1.23 - 1.96)	48 (1.62)	27 (0.28)	- -
Unmetamorphosed microgabbro (Devonian and Carboniferous ages only)	99	12 - 148 (1.08 - 2.17)	57 (1.69)	31 (0.25)	54 (15)
Metamorphosed microgabbro (Devonian and Carboniferous ages only)	69	4 - 121 (0.80 - 2.08)	46 (1.51)	33 (0.39)	72 (26)

tion B is within the range of the mean copper contents of the above groups of rocks which vary from 60 PPM to 80 PPM (Table 1).

Variation in Copper with Age.

It is evident from Table 1 that there is no real difference between the copper contents of the microgabbro of lower Devonian (60 ± 34 PPM), middle Devonian (61 ± 26 PPM) and upper Devonian (63 ± 29 PPM) ages. The same is true for the copper content of the Carboniferous microgabbro (60 ± 34 PPM) as compared to those of the Devonian. The Ordovician microgabbro however seems to have higher mean copper content (76 ± 35 PPM) which when subjected to t-test against the combined mean copper content (62 ± 30 PPM) for the rocks of the three divisions of the Devonian period apparently belonging to the same population, indicates that the difference is not significant. Therefore, it is concluded that the data at hand do not show any real variation in the copper content of the microgabbro of different ages.

Among the extrusive rocks, the pillow lava of Carboniferous age has significantly higher copper content (65 ± 34 PPM) compared to the pillow lavas of upper Devonian age (35 ± 17 PPM) as indicated by t-tests on parameters calculated from log-transformed values ($t = 3.10 > t_{.95} = 1.7$ for $v = 25$) and also in relation to the Permian basalts (48 ± 27 PPM) on the same basis ($t = 2.20 > t_{.95} = 1.77$, $v = 13$). The copper content of the Permian basalt is, however, not significantly different from that of the pillow lava of upper Devonian age.

Variation in Copper with the Type of Rocks

The distribution of copper in the various types of rocks has been summarized in

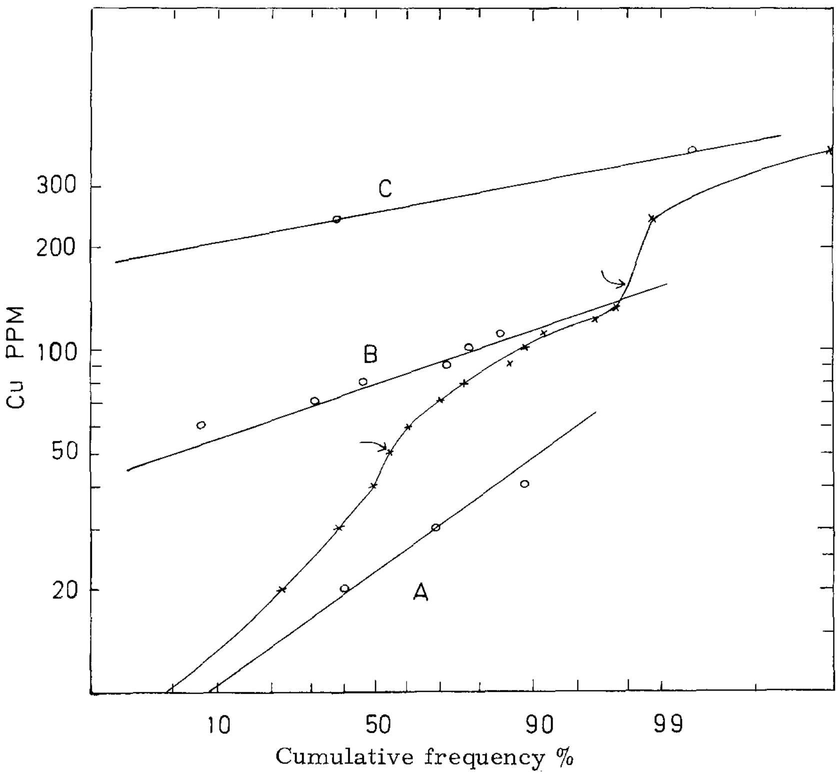

Fig. 3. Cumulative frequency plot and separated populations for the metamorphosed mafic rocks. As in Fig. 2, the curve passing through the crosses(based on 81 results) has the form of three populations on the basis of inflection points at 55.50 and 97.50 cumulative percentiles. The separated populations in the ratio 55.50/42.00/2.50 are represented by lines A, B and C

Table 2. The microgabbro as a whole seems to have relatively higher mean copper contents (59 ± 32 PPM) as compared to the picrite (38 ± 7 PPM), proterobase (50 ± 20 PPM) and quartz microgabbro (41 ± 10 PPM) but the differences between the above group of rocks do not seem to be significant as indicated by t-tests on transformed values. Among the extrusive rocks, the spilitic lava flows and the Permian basalt are found to contain 43 ± 27 PPM and 48 ± 27 PPM respectively and the difference between the two means is not significant.

However, when comparison is made between the copper contents of the intrusive microgabbro and extrusive pillow lavas representing the two major categories of rocks it is found that the mean copper content of the microgabbro (59 ± 32 PPM) and that of the pillow lava (43 ± 27 PPM) is statistically significant ($t = 2.33 > t_{.95} = 1.65$, $v = 159$).

Effect of Metamorphism

As in case of the unmetamorphosed mafic rocks, the frequency distribution in metamorphosed mafic rocks showed considerable positive skewness and therefore a cumulative plot was prepared on a log-probability paper. Because of the presence of inflections, the data were separated into three populations A, B and C (Fig. 3) with mean values of 23 PPM, 80 PPM and 250 PPM.

The population distribution in both the metamorphosed as well as unmetamorphosed mafic rocks appears to be similar except that there appears to be suggestion of certain amount of redistribution of copper in the metamorphosed rocks as evidenced by higher coefficient of variation (72 %) as compared to that (54 %) for the unmetamorphosed microgabbro. Further also as can be seen from Figs. 2 and 3, the metamorphosed microgabbro has a lower mean value (23 PPM) than the mean (33 PPM) for population A of the unmetamorphosed rocks, suggesting that copper has been dispersed and mobilized under the influence of intense contact metamorphism. Looking at the actual data, it is seen that the mean copper contents of the metamorphosed microgabbro of different ages are found to be lower than those of the unmetamorphosed microgabbro of the corresponding ages (Fig. 4). The difference in each case is small but nevertheless consistent. Therefore, combined means for the metamorphosed microgabbro (Devonian and Carboniferous) and unmetamorphosed microgabbro

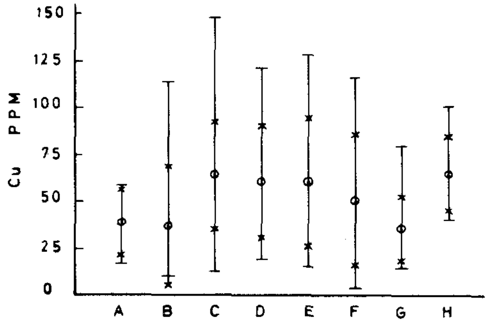

Fig. 4. Variation in the copper content of the unmetamorphosed and metamorphosed mafic rocks of various ages: (A) unmetamorphosed and (B) metamorphosed microgabbro of unclassified Devonian age; (C) unmetamorphosed and (D) metamorphosed microgabbro of upper Devonian age; (E) unmetamorphosed and (F) metamorphosed microgabbro of Carboniferous age; (G) unmetamorphosed and (H) metamorphosed pillow lava of upper Devonian age.
Symbols: 0 = mean, x = standard deviation, ⊢—⊣ = range

of corresponding ages have been calculated to be 46 ± 33 PPM and 57 ± 31 PPM and the difference is found to be significant ($t = 2.90 > t_{.95} = 1.65$, $v = 166$). The contact metamorphism of microgabbro north of Land's End granite is known to have resulted in large scale migration of elements (Tilley, 1935). The similarity in the mean copper content of the population B of the metamorphosed as well as unmetamorphosed microgabbro perhaps suggests that in many cases, particularly where the contact metamorphism may not have been intense, the distribution of copper has remained relatively unaffected. Population C of the metamorphosed microgabbro perhaps represents the presence of mineralized population although not well defined again due to the sampling bias. However, it is interesting to note that the presence of the mineralized population is indicated at a lower level (> 130 PPM) as compared to the same in the unmetamorphosed mafic rocks (> 150 PPM). This again suggests dispersion of copper from the metamorphosed microgabbro.

The pillow lavas which are generally porous and lie farther away from the granitic contacts seem to indicate an increase in copper content. For example the mean copper content of the Devonian metamorphosed pillow lava (64 ± 20 PPM) has increased by 29 PPM as compared to that in the unmetamorphosed variety (35 ± 17 PPM) as shown in part of the Fig. 4 and is statistically significant ($t = 2.44 > t_{.95} = 1.70$, $v = 27$). The increase in copper content in pillow lavas is considered due to permeation of hydrothermal solution emanating from the adjacent granitic bodies. Field relations indicate that the pillow lavas are porous and generally situated at a considerable distance from the granitic bodies and have suffered mild metamorphism along with hydrothermal permeation.

SEDIMENTARY ROCKS

A total of 88 samples of sedimentary rocks of different types and ages have

Table 3. Summary of the abundance of copper in the sedimentary rocks

Rocks		Results	Range PPM	Mean PPM	Standard deviation	Coefficient of variation (%)
Unclassified Devonian	Carbonaceous grey slates	11	30 - 87	45	19	42
	Noncarbonaceous grey slates	8	11 - 56	24	15	62
Lower Devonian	Carbonaceous grey slates	7	28 - 60	44	10	23
	Noncarbonaceous grey slates	7	5 - 73	31	23	78
	Noncarbonaceous purple slates	8	3 - 13	6	3	50
Middle Devonian	Carbonaceous grey slates	7	26 - 60	44	11	25
	Limestones	6	3 - 15	8	4	50
Upper Devonian	Carbonaceous grey slates	2	74 - 78	76	-	-
	Noncarbonaceous grey slates	6	7 - 53	34	18	53
	Noncarbonaceous purple slates	4	7 - 19	14	5	36
Carboniferous	Carbonaceous grey slates	15	6 - 56	28	13	46
	Metamorphosed slates	5	11 - 91	48	30	63

been analyzed for their copper contents (Table 3). In general the unmetamorphosed grey slates contain copper ranging from 5 to 87 PPM about 57 % of the results revealing copper content less than 40 PPM. In contrast to this, the purple slates have copper content ranging from 3 to 19 PPM while the limestones contain 3 to 15 PPM copper. Out of the total number of analyses only two yielded copper content more than 100 PPM. These are 205 PPM (Unclassified Devonian) and 224 PPM Carboniferous metamorphosed slate) and have been excluded from the calculation of mean and standard deviation for the respective groups because these are considered to represent mineralized population as discussed below.

Frequency Distribution

The cumulative frequency distribution based on the results of all unmetamorphosed slates (76 results) was plotted both on arithmetic and log probability graph papers to see which one gave a more linear plot. As the distribution showed only moderate positive skewness and a more linear plot was obtained on arithmetic probability paper, it was inferred that the distribution of copper in the sedimentary rocks was more nearly normal rather than lognormal. It is noted that the normal distribution of copper in this case is perhaps an exception to the rather general rule of lognormal distribution of trace elements in rocks as recognised by Ahrens (1953, 1954a, 1954b and 1957).

The cumulative frequency plot in this case as well showed inflection suggesting mixing of populations. Therefore, the cumulative frequency plot was partitioned into two populations A and B with mean copper contents of 29 PPM

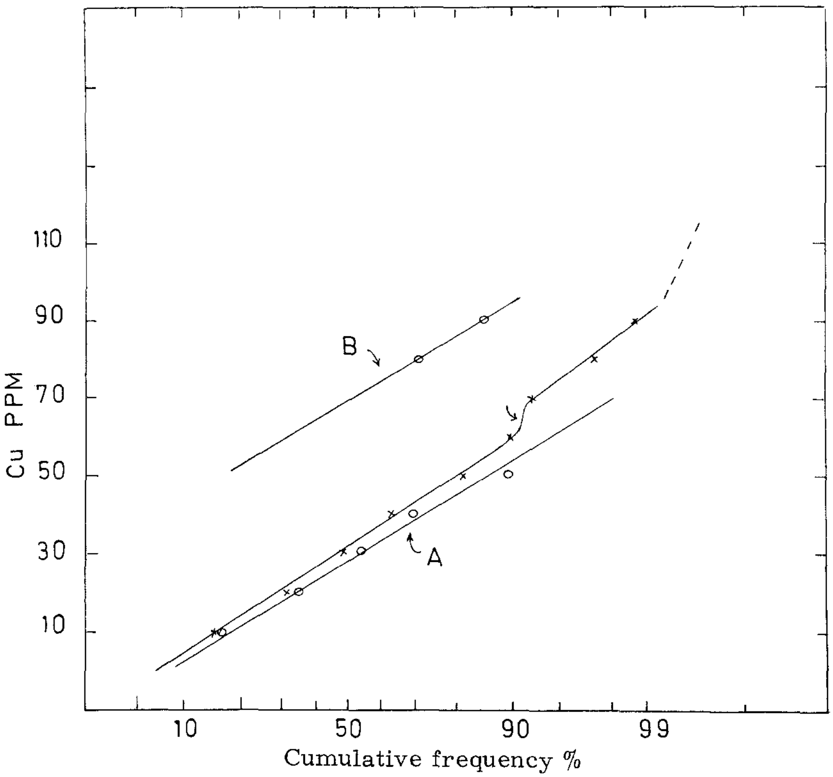

Fig. 5. Cumulative frequency plot and separated populations for the sedimentary rocks. The curve passing through the crosses (bases on 76 results) has the form of at least two populations based on an inflection at 91.0 cumulative percentile. The upper end of the curve represented by dashed line indilation which is very poorly represented in the data and therefore ignored. The two partitioned populations in the ratio 91.0/9.0 are estimated by the lines A and B

and 70 PPM respectively (Fig. 5). A third mineralized population with copper content more than 100 PPM is present but is very poorly defined and therefore, its presence has been ignored. It seems that the population A is represented by the noncarbonaceous slates, generally having lower mean values, while the population B is represented by the carbonaceous slates having higher mean values.

Variation in Copper with Age

The carbonaceous slates of the various divisions of Devonian period do not vary much in their copper contents but the Carboniferous slates seem to have relatively lower copper content. Therefore, a combined average for all the carbonaceous slates of Devonian period (27 results) has been calculated to be 47 ± 16 PPM and is found to be significantly higher than the mean copper content (28 ± 13 PPM) of the carbonaceous slates of Carboniferous age ($t = 3.8 > t_{.95} = 2.02$, $v = 38$). The difference between the mean copper contents of the noncarbonaceous slates of unclassified Devonian (24 ± 15 PPM), lower Devonian (31 ± 23 PPM) and upper Devonian (34 ± 18 PPM) is very small and is not significant.

Variation in Copper with Type

The distribution of copper in the different types of rocks has been shown in

Fig. 6 which indicates that the differences in the mean copper contents of the three different types of slates are significant because in each case the mean value lies beyond the standard deviations for the others. Copper content in the limestone is significantly lower as compared to that in the carbonaceous as well as noncarbonaceous slates.

Effect of Metamorphism

Only six samples of metamorphosed slates, all obtained from quarries north of Dartmoor, have been analyzed which may therefore give a biased estimate. The average copper content in these slates has been found to be 48 ± 30 PPM and is higher than the mean copper content of 28 ± 13 PPM for the unmetamorphosed slates of Carboniferous age as indicated by a t-test. ($t = 2.27 > t_{.95} = 1.73$, $v = 10$). The increase in copper content in the metamorphosed slates is apparently due to permeation of hydrothermal fluid emanating from the granitic mass.

GRANITIC ROCKS

A total of 78 samples representing coarse granite, fine granite, aplite, pegmatite and felsite have been analyzed (Table 4). The results of the analyses immediately reveal that there is a strong regional difference in the distribution of copper in the granites. The granites of Land's End and Carnmenellis masses are generally much richer in copper compared to those of St. Austell, Bodmin Moor and Dartmoor.

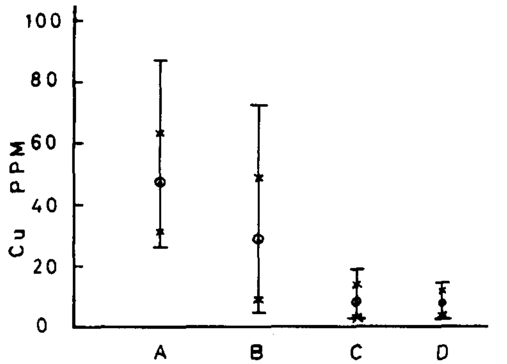

Fig. 6. Variation in the copper contents of the different types of sedimentary rocks: (A) all carbonaceous slates, (B) all noncarbonaceous slates, (C) all purple slates and (D) limestones. (Symbols same as in Fig. 4)

Frequency Distribution

When the cumulative frequency distribution of copper in all the granitic rocks (78 results) is plotted on an arithmetic probability graph paper (Fig. 7) it is interesting to note that the distribution can be approximated by two subparallel straight lines. This suggests that the data represent two separate populations with an intermediate zone caused by

Table 4. Summary of the abundance of copper in the granitic rocks

Rocks		Results	Range PPM	Mean PPM	Standard deviation	Coefficient of variation (%)
Land's end and carnmenellis	Coarse granite	19	21 - 108	76	16	21
	Fine granite	4	53 - 91	71	17	24
	Aplite	7	11 - 91	62	26	42
St. Austell, Bodmin Moor & Dartmoor	Coarse granite	25	5 - 30	17	8	47
	Fine granite	9	15 - 33	24	5	21
	Aplite	4	25 - 29	27	2	7
	Pegmatite	5	21 - 49	30	13	43
	Felsite	5	16 - 47	27	12	44

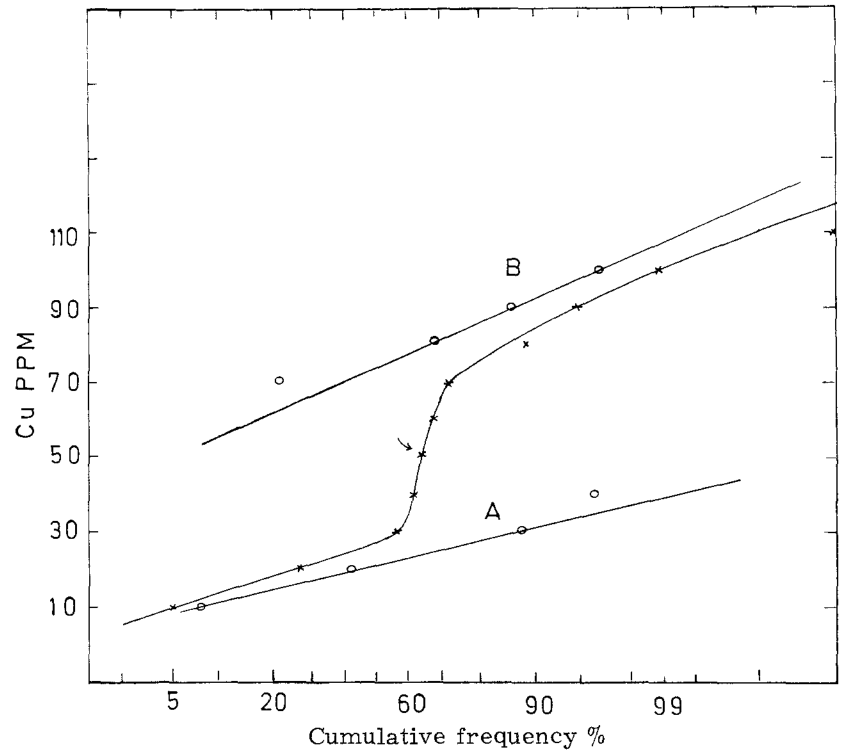

Fig. 7. Cumulative frequency plot and separated populations for the granitic rocks. The curve passing through the crosses (based on 78 results) reveals two segments of subparallel straight lines strongly suggesting that it represents a combination of two populations. The near vertical segment representing the central portion of the curve is presumed to be due to overlapping of the two populations. The point of inflection at 64.0 cumulative percentile at the centre of the mixed zone has been taken for the separation of the two populations. The two partitioned populations in the ratio 64.0/36.0 are estimated by lines A and B

overlapping of the two populations. A frequency histogram of the data (not shown) also results in two sharply differentiated portions each symmetrically distributed with widely different modal classes. The population has therefore been separated into two. When we look at the copper values from the different granitic masses, it becomes immediately apparent that the Land's End and Carnmenellis masses have yielded much higher values than those from the St. Austell, Bodmin Moor and Dartmoor masses and represent population A and B respectively. Therefore the following discussion is based on the above mentioned grouping.

Copper in the Land's End and Carnmenellis Masses

Altogether 30 samples from these two granitic bodies have been analyzed of which 19 represent coarse granite, 4 fine granite, and 7 aplite containing mean copper contents of 76 ± 16 PPM, 71 ± 17 PPM and 62 ± 26 PPM copper respectively. The variation in the copper content within these groups is not significant.

Copper in the St. Austell, Bodmin Moor and Dartmoor

A total of 48 samples of the granitic rocks from these masses have been analysed for their copper contents (Table 4). The values for mean, range and standard deviation for the distribution of copper in these rocks are represented in Fig. 8. The mean copper contents in the rocks ranging from the early coarse granite to the late pegmatite becomes consistently higher but in felsite copper content slightly decreases. A variance test was carried out to see if the difference within this group of rocks was significant. The calculated variance ratio 4.36 for $v_1 = 4$ (greater variance) and $v_2 = 43$ (lesser variance), is greater than the critical value of F at 5 % level (F = 2.4). Hence it is suggested that the samples were drawn from sources which differed in their copper contents. Therefore, it is considered that there has been a general enrichment of copper towards the pegmatitic stage.

SUMMARY

Mafic Igneous Rocks

The study has revealed that among the mafic rocks of the southwest of England, the mean copper content of the intrusive microgabbro (59 ± 32 PPM) is higher than that for the spilitic pillow lavas (43 ± 27 PPM) .

The microgabbros of different ages do not vary significantly in their copper contents. The pillow lavas of Carboniferous age are found to have more copper (65 ± 34 PPM) than those of the upper Devonian age (35 ± 17 PPM). The picrites of relatively more mafic composition have less copper content (38 ± 7 PPM) than the average microgabbro (59 ± 32 PPM). A similar relationship has been observed by Wager and Mitchell (1953) in case of the Hawaiian lavas and by Wager et al. (1957) in case of the Skaergaard intrusion.

Metamorphism of the microgabbro has resulted in the mobilization and dispersion of copper and may have contributed to the enrichment of copper ores in the region.

The cumulative frequency plots of both unmetamorphosed and metamorphosed mafic rocks suggest a lognormal distribution and indicate the presence of mixed populations. Existence of a mineralized population in each case is also indicated.

The copper contents of the intrusive as well as extrusive mafic rocks of Cornwall and Devon are low as compared to the figures available for similar rocks from other parts of the world. Vinogradov (see Taylor, 1964) has reported an average of 100 PPM copper in the basaltic rocks of U. S. A., while Ahrens and Fleischer (1960) have given similar figure for the microgabbro W-I. Although values as low as 72 PPM for Ontario microgabbro (Fairbairn et al. 1953) and 87 PPM for basaltic rocks of U. S. A. (Turekian and Wedepohl, 1961) have been reported, these are still higher than those for the rocks of Cornwall and Devon. Some reported values such as 46 PPM copper in the basalt of Indian Ocean ridge (Zlobin and Dmitriyev, 1968), 63 PPM in the basalts from Kurile Island (Markhinin and Sapozhnikova, 1962), 56.1 PPM in the metagabbro of north western Ontario (Emslie and Holman, 1966) and 43 PPM in the basalts from Siberia (Nesterenko et al., 1964) are comparable to that found in the microgabbro of the Southwest England.

Sedimentary Rocks

The copper content of the slates appears to be related to the lithology. Thus the carbonaceous slates are found to contain higher copper content (47 ± 16 PPM) than the noncarbonaceous grey slates (29 ± 20 PPM). A similar relationship has been noted by Le Riche (1959) in case of the sediments of southern England where copper content is found to increase with the increasing carbon content in the material. The purple slates of the area, presumably deposited under oxidizing conditions, are still poorer in copper content (8 ± 5 PPM). Gorham and Dalway (1965) noted the enrichment of copper, sulphur, nickel and carbon in the organic mud as compared to the oxidate crust of some British lakes.

The grey slates of Cornwall and Devon contain a mean copper content of 37 ± 19 PPM (based on 63 results) and compare favourably with the figures available for argillaceous rocks from other areas of the world. Macpherson (1958) has reported 42 PPM copper in Precambrian argillaceous rocks, while Barnes (1957) has found 50 PPM copper in Paleozoic shales. Very few values as high as 65 PPM in shale and clays of Illinois (White, 1959), 65 PPM in Paleozoic marine shales of Europe (Morita, 1955) have been reported. Values as low as 33 PPM (Nicols and Loring, 1962) and 18 PPM copper (Shaw, 1954) have also been reported.

Granitic Rocks

The distribution of copper in the granitic rocks of the area appears to be related to the degree of mineralization. The granitic rocks from Land's End and Carnmenellis masses on the average contain more copper (73 ± 18 PPM) than those from St. Austell, Bodmin Moor and Dartmoor (21 ± 9 PPM). Records of copper production show that the areas surrounding the former two granitic bodies have been the richest copper producing region (Dines, 1956). Parry and Nackowski (1963) have reported that the biotites from the mineralized area are relatively rich in copper in relation to those from unmineralized area in the Basin and Range quartz monzonite, and Warren and Delavault (1960) found much higher copper in granitic rocks associated with copper ores.

The granitic rocks of the relatively less mineralized region contain on an average 21 ± 9 PPM copper (based on 48 results) which is relatively high as compared to average copper contents of granitic rocks from other areas of the world. Such values as 8 PPM in granites from northwestern Ontario (Emslie and Holman, 1966), 11.8 PPM in granites from Central Massif, France (Isnard, 1970), 8 PPM in granites from

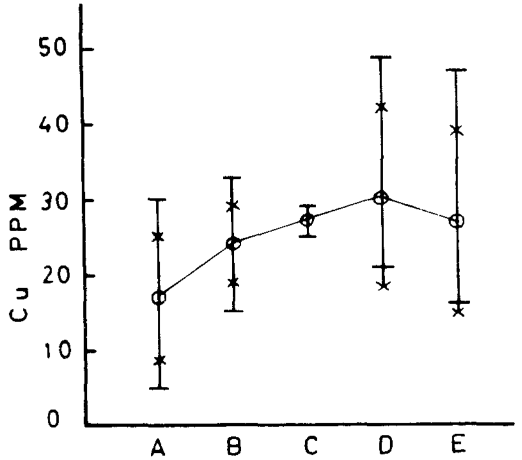

Fig. 8. Variation in copper content of the granitic rocks: (A) coarse granite, (B) fine granite, (C) aplite, (D) pegmatite and (E) felsite from St. Austell, Bodmin Moor and Dartmoor masses (Symbols same as in Fig. 4)

the Cape Province (Kolbe and Taylor, 1966) and 13 PPM in G-I have been reported. The average copper content for the granitic rocks from the highly mineralized region is 73 ± 18 PPM (based on 30 results) and is much too high for its class.

Relevance to Geochemical Exploration

The intention of the present work was primarily to study the abundance of copper in unaltered rock samples free from any apparent sign of mineralization and therefore, the results were not expected to be particularly relevant to geochemical exploration. However, the frequency distribution of copper in mafic rocks and particularly in granitic rocks shows that the study of primary dispersion of copper in the rocks may be of value in exploration work. The evidence that a mineralized population occurs at a lower level (130 PPM) in the metamorphosed mafic rocks than that in the unmetamorphosed mafic rocks (150 PPM) is a notable feature. Metamorphism of microgabbro has resulted in general mobilization and dispersion of copper leading to the decrease in its abundance and therefore it seems reasonable to expect that any presence of mineralization in the metamorphosed rocks will be revealed at a lower level.

The results of the distribution of copper in the granitic rocks reveal a more obvious applied aspect. As noted earlier, the cumulative frequency plot for copper in the granitic rocks could be immediately divided into two populations; population A having mean copper content 3.5 times as high as that for population B. Since it is known that copper production from areas surrounding Land's End and Carnmenellis masses represented by population A was 10 times higher than the same from areas around the other granitic masses represented by population B, the higher mean copper content of population A becomes immediately significant.

Therefore, it is considered that geochemical exploration based on rock analysis can provide important guides in the search and location of mineral deposits. Warren and Delavault (1960) recognised productive and unproductive granites as a result of aqua regia extraction of copper and zinc from granitic rocks scattered over 50,000 square miles and suggested that exploration based on rock analysis could be an important method in areas of scanty exposures and poor soil development.

ORIGIN OF THE COPPER DEPOSIT

A great deal has been written about the genesis of the ore deposits in Southwest England particularly with reference to the processes of mineralization and sequence of ore deposition (Dines 1934, Hosking 1949, 1951). On the other hand very little has been said about the source of metallic elements themselves. Probably it has been tacitly assumed that the initial granitic melt was rich in all the metals. As the differentiation of the magma proceeded the residual liquid became rich in copper and other metals which were finally deposited hydrothermally.

Several lines of evidence, including the results of present study, as discussed below suggest that the source of copper may not lie in the granitic magma itself.

The Granitic Magma as a Source

It is known that rocks of granitic composition can originate in one of several ways such as i) differentiation of a parental basaltic magma, ii) by processes of metasomatism and iii) by palingenesis of the sialic crust and/or deep seated geosynclinal sediments.

The high level, cross cutting granitic batholith of Southwest England is assumed to have originated as a result of partial melting of sialic crust and to have been emplaced by a combination of processes such as assimilative granitization and forcible injection, etc. It may be pointed out that an independent magma source for the granites is favoured by Brammal and Harwood (1932) and Bott et al. (1958). Granted that the granites originated by partial melting of the sialic crust then it is unlikely that the original silicate melt would have had a high copper content because of the chalcophile character of copper. Copper content of the sialic crust is not known. However, from the chemical nature of the sialic rocks as indicated by Daly (1933), it is presumed that the average copper content would not be much higher than 15 PPM, and possibly even lower as suggested by average values for granitic rocks from many parts of the world. Such a low copper content of the parent material is unlikely to furnish sufficient copper to account for the high values in the two granites and its subsequent concentration in the hydrothermal fluids depositing a huge quantitiy of copper ore. This becomes more unlikely in view of the fact that the present study indicates relatively higher copper content (21 PPM) than normal even in the granites of the less mineralized region. Therefore, it seems that the source of copper does not lie in the parent material but perhaps somewhere else.

Table 5. Volume occupied by the granitic bosses (area measured by planimeter)

Granite bosses	Area sq. mile	Total area sq. mile	Total miarea sq. km	Volume km^3 (thickness 15 km)*	Total volume km^3
Land's End	73.20				
Carnmenellis	50.08	128	331.52	4973	
Godolphin					
					18726
St. Austell	32.20				
Bodmin Moor	78.40	354	916.86	13753	
Dartmoor	243.84				

*The thickness of 15 km is based on the estimate of Bott et al., 1958

Country Rocks as a Source

The country rocks of the region are largely composed of pelitic geosynclinal sediment and associated mafic rocks and have been found to contain average copper contents of 30 PPM (based on 81 results) and 58 PPM (based on 180 results) respectively. The country rocks may have acted as a source of copper in two different ways. One of the ways of contribution from the country rocks is considered dependent on the extent of assimilation of these rocks themselves during the evolution of the granitic magma. That the process of assimilative granitization of country rocks may play an important role in the production and evolution of granitic magma is generally recognized (Bowen 1956, Wilson 1938, Goodspeed 1952, Khitarov and Pugin 1964). Lacy (1960) has indicated that about 25 to 30 % of the material making up a batholith may be derived as a result of assimilation of existing geosynclinal rocks during its rise to upper crustal level.

Assuming that 25 % of the magma forming the granitic rocks of the southwest of England has been derived as a result of assimilation of the country rocks then calculations indicate (Table 5) that about 4681.50 km^3 of granitic rocks represent assimilated country rocks. Assuming a volumetric ratio of 1:25 between the mafic rocks and the sedimentary rocks (in fact areal measurement of surface exposures by planimeter suggests a ratio of 1:23), it would mean that about 187 km^3 of mafic rocks and 4494 km^3 sedimentary rocks were assimilated. It can be easily shown that $1 \times 10^6 m^3$ [1000m x 1000m x 1m] of mafic rocks of density 2.9 with an average copper content of 58 PPM [2] will have 168 tons of copper. Therefore, 187 km^3 or $187 \times 10^9 m^3$ of mafic rocks supposed to have been assimilated in the granitic magma would have contributed about $187 \times 10^3 \times 168 = 31416 \times 10^3$ tons of copper. Similarly, 4494 km^3 or $4494 \times 10^9 m^3$ of sedimenatry rocks of density 2.7 with an average copper content of 30 PPM would have contributed about $4494 \times 10^3 \times 30 \times 2.7 = 364\,014 \times 10^3$ tons of Copper. Therefore, a total of $31\,416 \times 10^3 + 364\,014 \times 10^3 = 395\,430 \times 10^3$ tons of copper may have been contributed to the granitic magma as a result of the assimilation of the country rocks. This is about 500 times greater than the quantity of copper recovered as ores from the southwest of England which amounts to a total of about 798 704 tons of copper. Obviously all the copper thus contributed to the magma may not have been available for deposition as hydrothermal lodes. Considerable quantity of copper must have been retained by the granitic rocks in silicate structure or disseminated sulphide of no economic

2) Average copper content (58 ± 38 PPM) based on 180 results including all the microgabbro and spilitic lava.

significance. Such a quantitave treatment may be subject to criticism particularly in view of the fact that opinion may vary on the extent of assimilation of country rocks in the evolution of granitic magmas in general. However, the above calculation, tentative as it may be, indicates that considerable quantity of copper as well as other elements can be contributed to the granitic magma as a result of assimilation and incorporation of country rocks.

Contribution due to Circulating Water

In recent years, data have accumulated which attach considerable significance to the role of deeply penetrating ground water and its circulation in the process of enrichment and deposition of ores. White (1968, 1974), Skinner and Barton (1973) have more recently discussed and summarized origins of hydrothermal ore fluids. Isotopic studies have revealed that much interaction of meteoric water with magmatic water can take place particularly in shallow intrusives (Taylor 1971). Similar mixing of ground water with magmatic waters is indicated in many porphyry copper deposits such as Butte, Montana, Bingham, Utah, and Ely, Nevada (Sheppard et al., 1969 and 1971). Intrusion of hot igneous magma in otherwise relatively cool country rocks eventually sets up a circulation pattern of water and consequent leaching of metals from a zone of contact metamorphosed rocks and their inward migration can be important (Boyle 1959, Burbank and Leudke, 1968).

Therefore, it is considered quite likely that the probable circulation of meteoric and metamorphic waters during the emplacement of the granitic batholith of the southwest of England may have picked up copper from the surrounding country rocks and contributed to the hydrothermal fluids depositing the copper ores. Such a process may have contributed additional copper as distinct from the contribution due to direct assimilation of the country rocks.

Relevance of Isotopic Data

Brown (1962, 1965) has pointed out that the abundance of Pb 204 varies from 1.400 % (in the oldest rocks) to 1.340 % (in the youngest rocks) in all areas where lead occurs in the Phanerozoic. In a more recent survey of the ore lead isotopes of the British Isles, Brown (1966) pointed out that there is no lead in the range of 1.340 to 1.360 %. Most values including those from Devon (1.381 %, area No. 13) and Cornwall (1.376 %, area No. 12) are above 1.360 % and indicate generation from normal marine sediments. Further also, sericite from Cornwall indicates relatively D-rich water which perhaps suggests a large connate water component during the formation of sericite (Sheppard et al. 1971).

In summary, it may be said that the above discussion together with the relative abundance of copper in the country rocks in relation to those found in the granitic rocks seem to indicate that the copper, perhaps other elements too, deposited as ores were probably derived from the country rocks.

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