Thermophysical properties of the Lipari lavas (Southern Tyrrhenian Sea)

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Abstract

Results of thermophysical investigations into the lavas of the island of Lipari (Southern Tyrrhenian Sea) are presented. Samples selected for laboratory measurements belong to four main magmatic cycles, which produced basaltic-andesitic, andesitic and rhyolitic lavas. The wet-bulk density and the thermal conductivity measured on 69 specimens range from 1900 to 2760 kg m^{-3} and from 1.02 to 2.88 W m^{-1} K^{-1}, respectively. Porosity is never negligible and its influence on density is maximum in rhyolites of the third cycle. The thermal conductivity is also influenced by the amount of glass. Rhyolitic obsidians show values lower than other rhyolites, although the latter rocks have a larger average porosity. The radioactive heat production determined on 36 specimens varies with the rock type, depending on the amount of U, Th and K. In basic lavas of the first cycle its value is 0.95 ± 0.30 μW m^{-3}, while in rhyolites of the fourth cycle it attains 6.68 ± 0.61 μW m^{-3}. A comparison between results of γ-ray spectrometry and X-ray fluorescence points out that the assumption of equilibrium in the decay series of the isotopic elements seems fulfilled. The information obtained is useful not only for the interpretation of geophysical surveys but also for the understanding of the geochemical characteristics of lavas.

Key words *Aeolian Islands – volcanic rocks – index properties – thermal conductivity – radioactive heat production*

1. Introduction

The petrogenetic and geophysical implications of the radiogenic heat sources in the recent volcanics of Southern Italy were discussed in a pioneer work by Civetta and Gasparini (1973). These authors demonstrated that the occurrence of magmatic products with very high concentrations of radioactive elements is extremely important in terms of geothermal balance. In the present paper we report the results of laboratory measurements of radioactive heat production and other physical parameters of geothermal interest carried out on volcanic rocks.

We focus on the island of Lipari, the largest of seven volcanic islands forming the Aeolian archipelago, located on the southeastern corner of the Tyrrhenian Sea (fig. 1). A chemical and mineralogical study was also made in order to discuss the physical properties in relation to rock composition, and assess the presence of mineral phases which could act as a reservoir of heat-producing elements. Determinations of U, Th and K concentrations were made by means of two independent techniques (γ-ray spectrometry and X-ray fluorescence). This allowed us to verify whether conditions of secular equilibrium occur in the radioactive decay series.

Lipari, together with the other Aeolian Islands and several submarine volcanoes, whose products consist of calc-alkaline, high-K calc-alkaline, shoshonitic and potassic series, aged upper Pleistocene-Present, is part of an arc-

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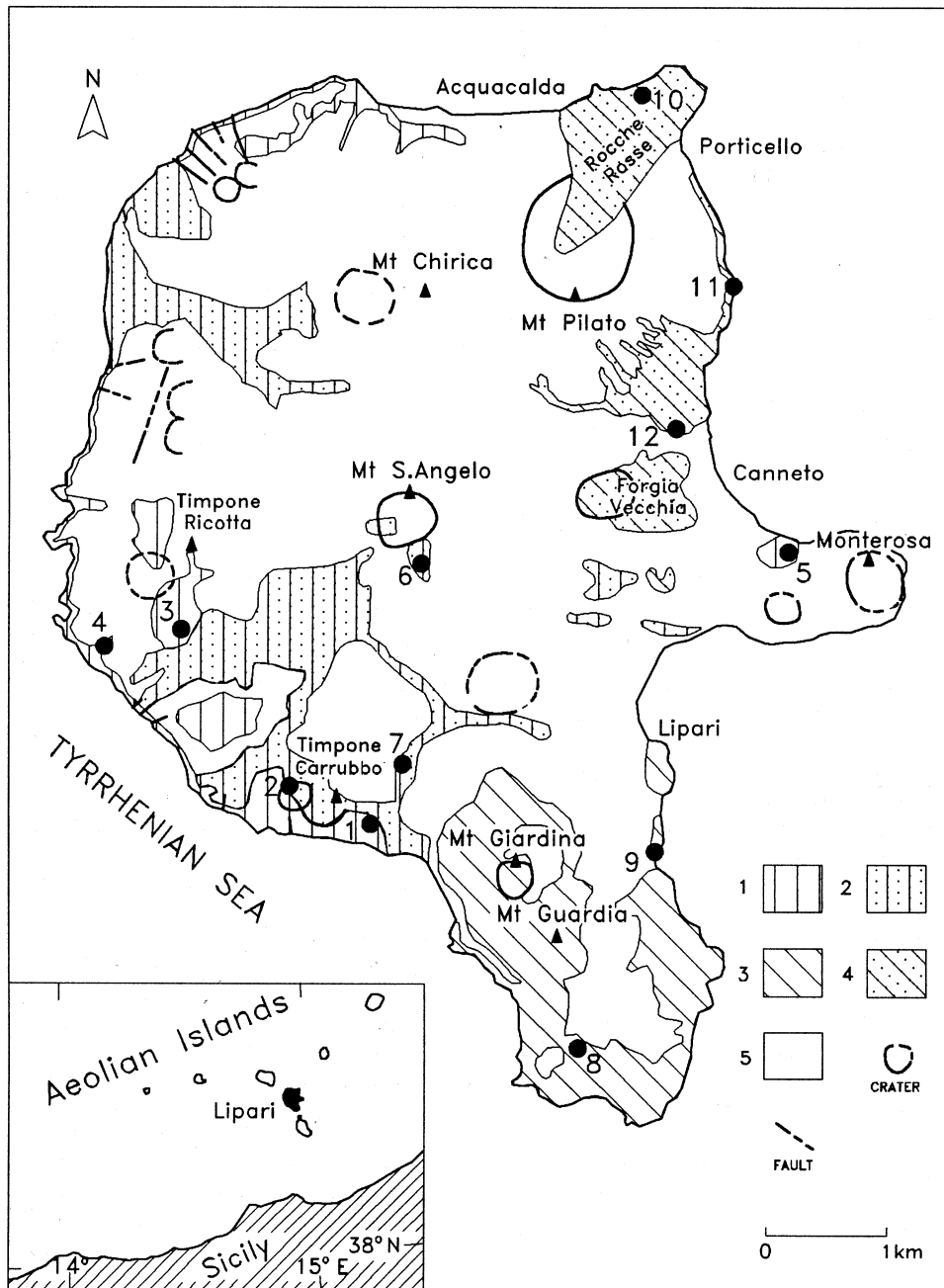
Fig. 1. Geological scheme of the island of Lipari and location of sampling sites (1-12). 1) Basaltic-andesitic lavas (cycle I); 2) andesitic lavas (cycle II); 3) rhyolitic lavas (cycle III); 4) rhyolitic obsidians (cycle IV); 5) pyroclastic deposits and pumices (all cycles).

shaped structure which borders the inner side of the Calabrian arc (Crisci *et al.*, 1991). Built up by tens of emission centres, often monogenic, Lipari has had a typically Strombolian activity, eruptive and explosive, with andesitic-basaltic products dated 223 ky, and more recently a phreato-magmatic activity with rhyolitic products (Pichler, 1980). At present it is quiescent, but shows low-temperature fumaroles and thermal springs.

The structural pattern is defined by a NW-SE trending fault system from which several N-S to NE-SW trending normal faults splay out, defining the major depression of the island (Mazzuoli *et al.*, 1995). The available volcano-tectonic and petrological information allows a systematic study of the evolution of the volcanic products with time. The variability of rock types is large, and their regional and temporal distribution is well explained by a petrogenetic model which involves complex mixing of magmas derived from a deeper sub-alkaline source with alkaline magmas, generally more evolved, formed at shallower depths (Pichler, 1980).

A stratigraphic sequence consisting of four distinct magmatic cycles was recognised by Pichler (1976), and more recent studies have outlined in detail the evolution of eruptive centres within the main cycles (Esperança *et al.*, 1992). The first and oldest cycle (from 223 to 150 ky ago) is mainly represented by submarine basaltic lava flows and hyaloclastites, related to N-S trending eruptive fissures developed along the western side of the island, and basaltic-andesitic lava flows and pyroclastics, linked with the activity of several volcanoes (*e.g.*, Mt. Chirica and Monterosa) located on the northern and eastern parts of the island.

The second cycle is characterised by volcanics mainly erupted from the Mt. S. Angelo (from 127 to about 92 ky ago) and by lacustrine deposits. The former are high-K andesitic lava flows, some of which show clear evidence of magma mixing and incorporation of basement rocks, represented by the extrusion of «cordierite-bearing» lavas (Barker, 1987). The latter fill a large depression, which extends in the central part of the island, and consist of reworked surge products, alluvial fan deposits and volcanoclastic turbidites.

Lavas of the third cycle, ranging in age from 42 to 23.5 ky, outcrop along a N-S trending belt in the southern part of the island. The activity started with the emplacement of rhyolitic lava domes and pyroclastics (Mt. Guardia and Mt. Giardina).

During the last cycle (from about 8.6 ky to historical times) volcanic centres (Forgia Vecchia and Mt. Pilato) formed from which obsidian lava flows and pyroclastics were erupted. The youngest products of this cycle, whose magmas derive by partial melting of continental crust, were dated to 729 A.D. (Cortese *et al.*, 1986).

2. Analytical techniques

The main lava formations were sampled at 12 sites (fig. 1), paying attention to select unweathered rocks. The sampled rocks pertain to lava flows of the main cycles of volcanic activity and depict the evolutionary trend of the island volcanism. Several specimens were prepared for measurements of physical properties, and some of them were pulverised for chemical analyses.

A series of specimens were cut into cylindrical shape, 18 mm in diameter and about 15 mm in length. In order to keep the accuracy of the volume determinations better than 1.5%, surfaces of each specimen were smoothed over a revolving disc by means of fine abrasives. This made such specimens also suitable for thermal conductivity measurements.

Specimens were dried for 24 h in an oven at 110 °C, and then weighed. Subsequently, they were placed in a vacuum drier for 24 h before the final weighing. Specimens were weighed by means of a gimbaled digital balance. In order to calibrate the balance and to determine errors, some standards were weighed both before and after each specimen. Both the accuracy and repeatability resulted less than 0.02 g, yielding an error of less than 0.1%.

Porosity was calculated by dividing the difference between the weight in the wet and dry states by the volume. The wet weight was determined after immersing each specimen in water for 36 h. Porosity is thought to be obtained with a maximum error of 6%, consider-

ing that not all pores are actually filled by fluid. The bulk density of the specimen in the wet and dry states was also determined.

X-ray diffractometry analysis was carried out on the diamagnetic heavy fraction of materials with the aim of identifying the main accessory minerals which are characterised by high gamma activity. Major and trace elements were analysed by means of X-Ray Fluorescence (XRF) spectroscopy. Accuracy, monitored using international standards, is better than 10% for elements with concentrations above 10 ppm. For Cr, Ni and U the detection limit is 2 ppm.

The equipment implemented for measuring the thermal conductivity consists, substantially, of two cylindrical blocks of copper, whose thermal capacity is known, between which the rock specimen is interposed. When the experiment begins, the stack of elements has a uniform temperature gradient. Afterwards, the lower block is cooled by immersing it into a refrigerant mixture. The amount of heat removed in a given time interval from the upper block, acting as a heat source, is measured. Experiments were carried out at room temperature and pressure, and accuracy is estimated to be better than 5% (Pasquale, 1983).

The heat-producing elements (U, Th and K) were determined in the laboratory of the Geological Survey of Finland by means of γ-Ray Spectrometry (GRS). Measurements were carried out with a NaI(Tl) detector optically coupled to a photomultiplier and a multichannel analyser for collection and storage of the energy spectra. In order to improve the sensitivity, the background radiation was smoothed by shielding the scintillator with lead. Both the detection limit and the possible error of the equipment are about 1 ppm for U and Th and 0.1% for K. Samples were usually about 250 g in weight, and ground to fit the well-type crystal (Kukkonen, 1989).

3. Materials and results

Materials from each site (fig. 1) were selected on the basis of thin section, mineralogical and chemical analyses. Samples of the first cycle lavas (sites 1 and 3-5) are porphyritic basaltic andesites. They contain mainly clinopyroxene and plagioclase phenocrysts (from 35 to 65%) in a fine-grained groundmass. In addition, biotite and red-brown horneblende phenocrysts were occasionally found (site 1). Altered olivine relics were rarely found (site 3).

Samples belonging to the second cycle lavas (sites 2, 6 and 7) vary from hypohyaline andesite with cryptocrystalline (site 6) or glassy groundmass. They contain phenocrysts of plagioclase, ortho- and clinopyroxene; cordierite, sillimanite and garnet are present (sites 2 and 7).

Third cycle lavas (sites 8 and 9) are rhyolitic, characterised by low (< 2%) phenocryst abundance (plagioclase, K-feldspar and clinopyroxene) either in a spherulitic fine-grained groundmass welded by brown glass (site 8) or in a densely packed spherulitic ground (site 9), indicating hydrothermal alteration processes.

The last cycle of lavas (sites 10-12) consists of rhyolitic obsidians characterised by aphiric textures (phenocryst abundance < 1%) with rare spherulitic aggregates spread out in an isotropic glassy groundmass (site 10) or spherulitic, densely packed aggregates showing layering textures welded by glass (sites 11 and 12) with irregularly shaped vesicular cavities.

The chemical analysis by means of X-ray fluorescence allowed us to discriminate terms which can be referred petrochemically to the calc-alkaline and high-K calc-alkaline magmatic series (fig. 2). Rocks showing basaltic-andesitic composition belong to the calc-alkaline series and less frequently to the high-K calc-alkaline series. The andesitic rocks of the second cycle and rhyolites (third and fourth cycles) are characterised by a marked enrichment in K_2O, such that they all belong to the high-K calc-alkaline series.

All the analysed rocks show very low K_2O/Na_2O ratios, typical of the sub-alkaline series, with a marked increase in SiO_2 towards the most differentiated terms, and a weak increase in the Fe/Mg ratio. The TiO_2 content is low also in the less evolved terms, where Al_2O_3 is relatively high (16-20% in weight; fig. 3).
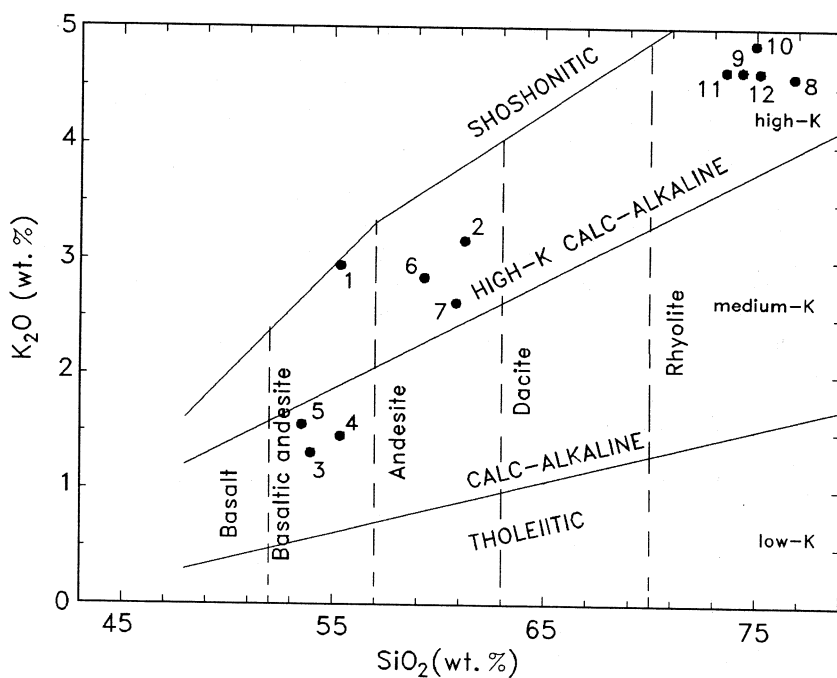
Fig. 2. K_2O *versus* SiO_2 data for lavas sampled at 12 sites of the island of Lipari superimposed on the classification by Peccerillo and Taylor (1976). The digit next to each point refers to the sampling site of fig. 1.
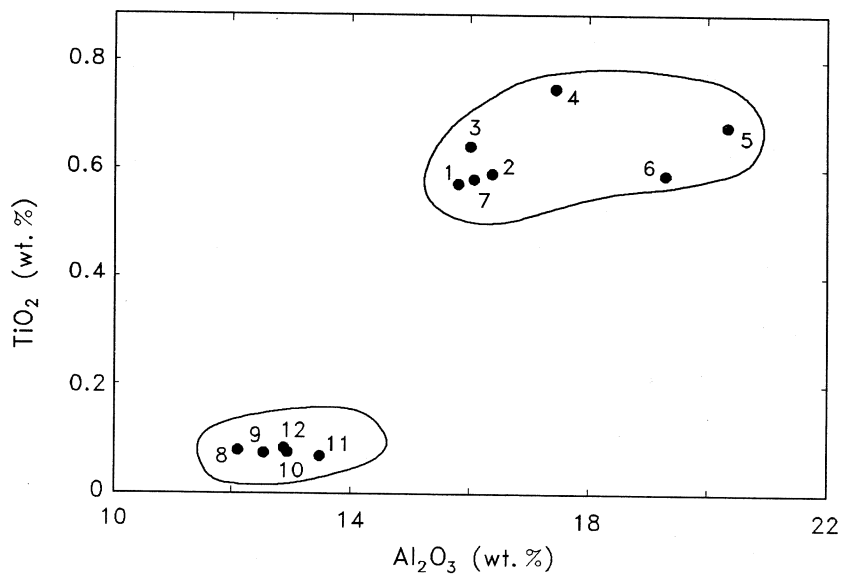

Fig. 3. A diagram of TiO_2 *versus* Al_2O_3. Details as in fig. 2.

A larger enrichment in large ionic radius elements, such as K, U, Th and Zr, is evident for the more evolved terms of the series. The distribution diagrams (fig. 4) show a clear positive correlation for all these elements *versus* SiO_2. In fact, extremely high Th and U concentrations occur in the rhyolitic rocks. The residence time in the mantle, the depth of origin and the interaction of a mantle-derived magma with the upper crust are important factors controlling the abundance of these elements within melts. Like K, U and Th are also contained in major minerals, but they occur in large concentrations only in a few, less abundant species, such as apatite, monazite, sphene and zircon. K is mostly locked up in feldspars and phyllosilicates (Rogers and Adams, 1969). Results of the X-ray diffractometry analysis in-

dicate that zircon is the only trace-mineral found in our samples.

Measurements of wet-bulk density, porosity and thermal conductivity were carried out on 69 specimens (at least 4 from each site). Figure 5 shows the distribution of the different sets of observations. The density varies from 1900 to 2760 kg m^{-3} while porosity from 0.5 to 28.2%. Their mean values are 2354 ± 207 kg m^{-3} and $11.3 \pm 6.6\%$, respectively. A wide interval of variation is observed also for the thermal conductivity, which ranges between 1.02 and 2.88 W m^{-1}K^{-1}.

In order to calculate the radioactive heat production, the abundance of U, Th and K was measured on 36 samples (3 for each site). All natural radioactive elements generate heat to a certain extent, but most of it (over 99%) de-
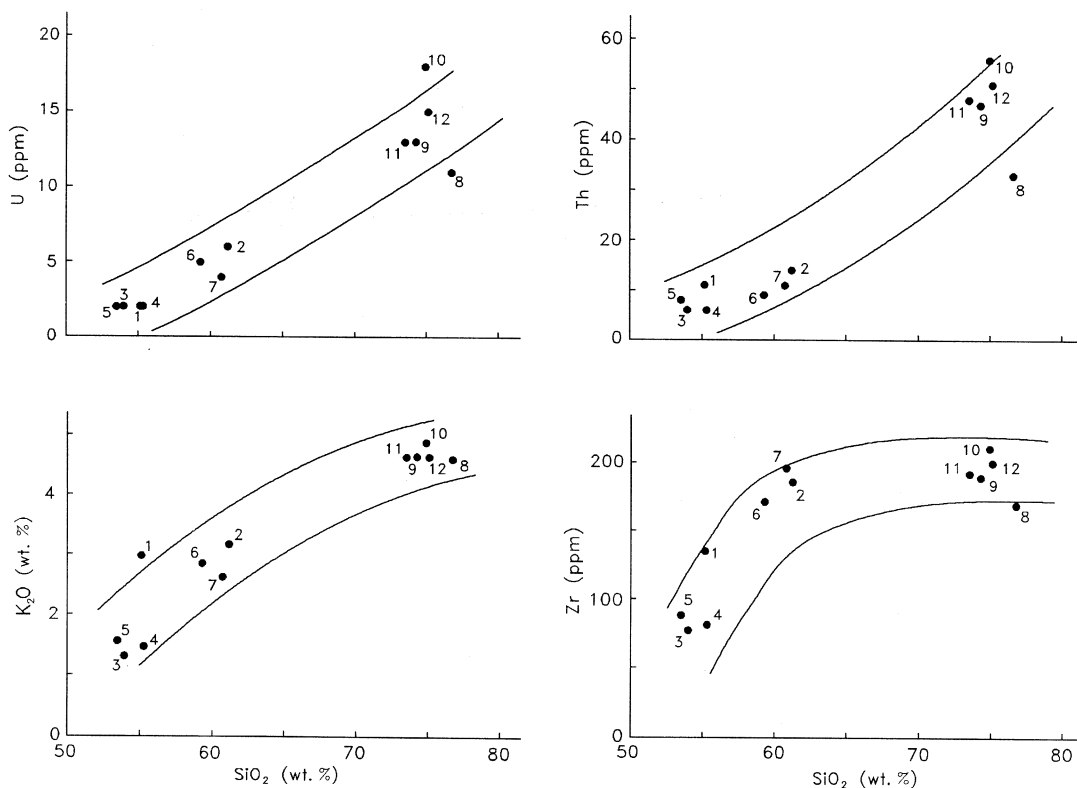

Fig. 4. Variations of U, Th, K and Zr *versus* SiO_2. Details as in fig. 2.
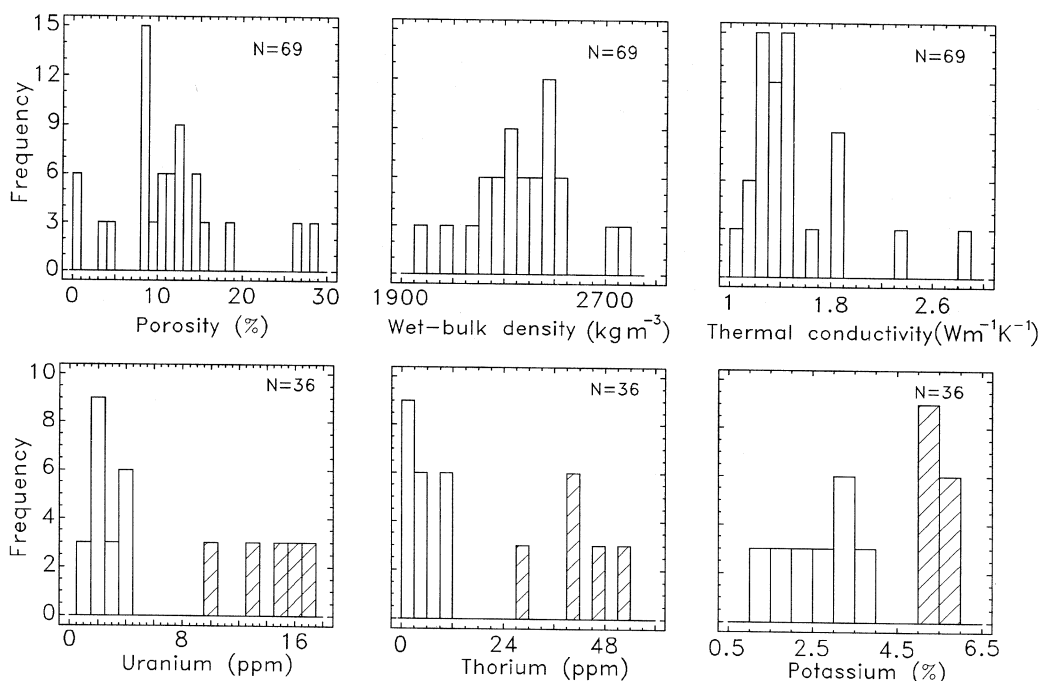
Fig. 5. Histograms of porosity, wet-bulk density, and thermal conductivity of 69 specimens of lava (above) and U, Th and K concentrations of 36 specimens (below). N is the number of samples. Hatching represents acid lavas.

pends on the amount of U, Th and K. The distribution of these elements in the examined rocks is shown in fig. 5. U and Th largely vary in concentration from 1 to 17 ppm and from 3 to 54 ppm, respectively. The largest values were found for the rhyolitic rocks of the third and fourth cycles. The Th/U ratio ranges from 1.5 to 3.2. The maximum concentration of K amounts to 6%.

On the basis of well known isotopic decay schemes, half lives and mass differences, we determined the radioactive heat production by means of the heat-production constants proposed by Rybach (1973), which give the heat generated for unit time and gram of U, Th and K. This property was expressed as the radiogenic heat-production rate per unit volume of rock. Figure 6 shows the histogram of the calculated values which range from 0.6 to 7.5 μW m^{-3}. The highest values found for the rhyolitic lavas.
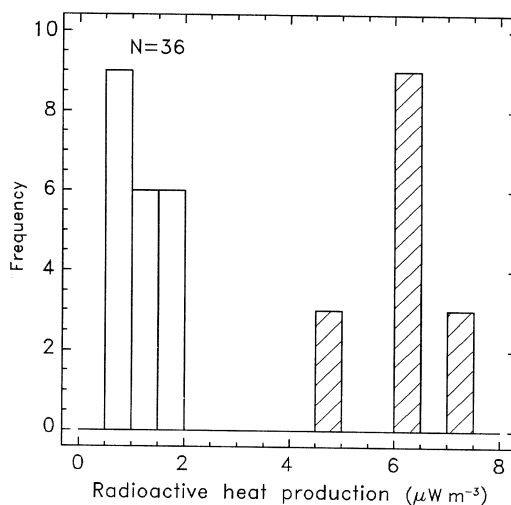

Fig. 6. Histogram of radioactive heat production values of 36 samples. Hatching represents acid lavas.

4. Discussion

Table I summarises the results of the basic index properties, the thermal conductivity and the radioactive heat production of the four main groups of lavas, together with the ranges of the SiO_2 and Al_2O_3 contents. Besides the wet-bulk density, we determined the grain density by dividing the dry weight of the specimen by the net volume excluding the pores. Under natural conditions, the wet-bulk density of rocks is affected by the mineralogical composition and porosity, whereas the grain density depends only on the composition. The grain density increases with the increase of Al_2O_3 against a decrease of SiO_2. The influence of porosity on the wet-bulk density is evident in the acid lavas (cycles III and IV), which has similar contents of Al_2O_3 and SiO_2.

The thermal conductivity has a more complex behaviour, as it decreases not only with porosity, but also with the increase in the glass

Table I. Average values and standard deviations (in brackets) of index properties, thermal conductivity, U, Th and K concentrations and radioactive heat production of lavas from Lipari. N is the number of specimens investigated. The range of SiO_2 and Al_2O_3 contents is also listed.

Rock type (volcanic cycle)	N	SiO_2 (wt. %)	Al_2O_3 (wt. %)	Porosity (%)	Wet-bulk density (10^3 kg m^{-3})	Grain density (10^3 kg m^{-3})	Thermal conductivity (W m^{-1} K^{-1})
Basaltic andesites (I cycle)	27	53-55	16-20	9.6 (3.9)	2.50 (0.16)	2.63 (0.11)	1.61 (0.52)
Andesites (II cycles)	18	59-61	16-19	12.9 (7.2)	2.36 (0.18)	2.56 (0.09)	1.27 (0.14)
Rhyolites (III cycle)	12	74-77	12-13	16.5 (5.9)	2.15 (0.16)	2.36 (0.10)	1.84 (0.38)
Rhyolitic obsidians (IV cycle)	12	73-75	13-14	7.3 (7.7)	2.24 (0.12)	2.34 (0.11)	1.28 (0.11)

Rock type (volcanic cycle)	N	U (ppm)	Th (ppm)	K (%)	Th/U	Radioactive heat production (μW m^{-3})
Basaltic andesites (I cycle)	12	2.0 (0.8)	3.8 (1.4)	2.1 (0.6)	2.0 (0.6)	0.95 (0.30)
Andesites (II cycles)	9	3.3 (1.0)	9.3 (2.7)	3.4 (0.2)	2.8 (0.3)	1.69 (0.38)
Rhyolitic (III cycle)	6	12.5 (2.7)	35.3 (6.0)	5.4 (0.0)	2.9 (0.2)	5.37 (0.76)
Rhyolitic obsidians (IV cycle)	9	15.3 (1.8)	47.5 (5.3)	5.7 (0.3)	3.1 (0.2)	6.68 (0.61)

content, which is characterised by a very low value (1.36 W m^{-1}K^{-1}) of thermal conductivity (Horai, 1971). Within acid rocks, rhyolitic obsidians of the fourth magmatic cycle show the smallest values of thermal conductivity (1.28 W m^{-1}K^{-1}).

GRS determinations were carried out on the assumption of secular equilibrium in the radioactive decay series. In a decay chain, under conditions of natural disintegration in an isolated system, radioactive equilibrium is reached for long time periods. Geologic processes (erosion, leaching, etc.) can separate parent and daughter isotopes, and some members could escape from the system, which would be seriously affected by disequilibrium. Whereas this hardly applies to the Th series, disequilibrium in the U chain can be present in weathered samples. No problem of such a kind arises from γ determinations of K (Rybach, 1988).

The field surveying revealed a widespread post-magmatic activity on the Lipari lavas which, at some outcrops, caused the total alteration of the original lithotype. This evidence strengthened the need for careful mineralogical analyses in order to assess to what degree these processes affect the condition of radioactive equilibrium, producing biased γ-spectrometry results. Although it was carried out on macroscopically unweathered, fresh samples, the thin section study allowed us to verify some evidence of alteration also at the mineral scale. The possibility that such phenomena affected to some extent also the groundmass, essentially consisting of microphenocrysts of plagioclase and glass, could not be excluded *a priori*, as the glass revealed evident devitrification. Besides, since the examined rocks are younger than 300 ky, strong radioactive disequilibrium should occur (Civetta and Gasparini, 1973).

Therefore, γ-spectrometry concentrations of U and Th were compared with direct determinations through X-ray fluorescence for a set of representative samples. A testing of the results of the two analytical techniques is shown in fig. 7. Within the limitations of the accuracy of both the measurement techniques, for U the two sets of results are well linearly correlated (correlation coefficient of 0.977). The intercept and slope of the line fitting the data are -0.659 ± 0.699 ppm and 1.042 ± 0.073, respectively.

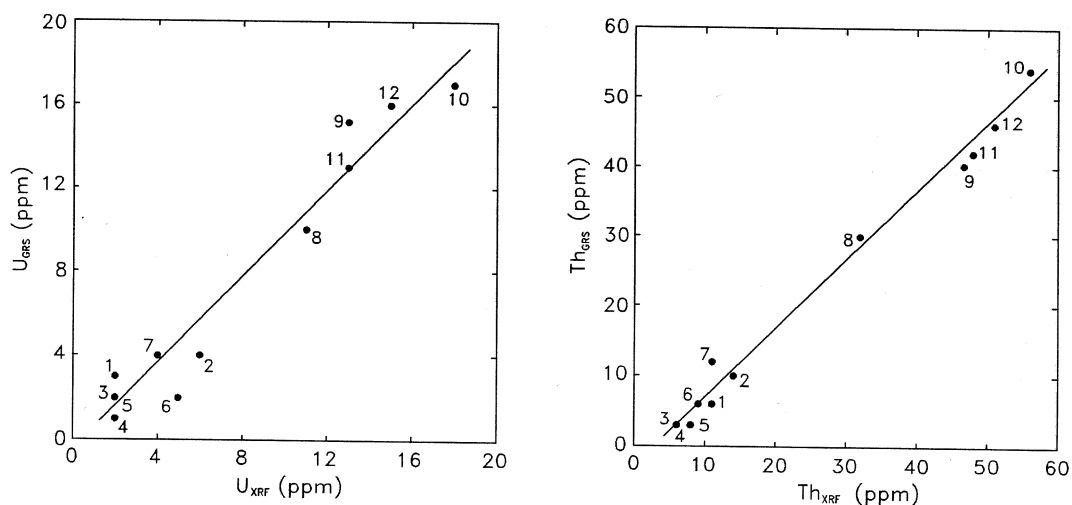**Fig. 7.** Linear correlation for U and Th concentrations from γ-Ray Spectrometry (GRS) *versus* X-Ray Fluorescence (XRF) results. Details as in fig. 2.

The γ-ray technique tends to underestimate the Th content. The line which best fits data has an intercept of -2.846 ± 0.954, slope of 0.970 ± 0.030, and correlation coefficient of 0.995. The underestimation is on average 10%, of the same order of magnitude of the analytical error in common rocks (Rybach, 1988). The good correspondence obtained for U and Th values is somehow in favour of secular equilibrium in the decay series of the investigated samples.

Radioactive heat production depends on the amount of U and Th trace elements and of K as a mineralizer in the rocks. Even though the concentration of these elements in each sample varies widely, it increases with the SiO_2 content. The U and Th abundance in the rocks of rhyolitic composition is on average about 5 times larger than that of basic lavas. This implies that the greater average value of radioactive heat production ($6.7\ \mu W\ m^{-3}$) was found in rhyolitic obsidians.

A comparison between our results of concentrations of heat-producing elements and those reported by Civetta and Gasparini (1973) and Francalanci et al. (1993) is possible for a few rocks. Values from earlier studies do not differ significantly from our γ-spectrometry determinations. However, it must be stressed that U, Th and K contents were inferred by means of different techniques (spark-source mass spectrometry, neutron activation analysis and isotope dilution α-ray spectrometry), and too few samples were analysed.

The binding mechanisms of U, Th and K result in two principal constituents, namely a readily soluble and a less soluble component. The less soluble part is fixed in the crystal lattice of one or more minerals, in inclusions or ionic exchange position incorporated or built into the mineral structure. The soluble part is that which is absorbed at grain boundaries or crystal surface and/or is found in pore space of a rock. This component becomes mobile as soon as water migrates through the rock. A redistribution especially of U, but also of Th, takes place not only during weathering but also during the cooling of intrusions in the upper crust (Rybach and Buntebarth, 1981). The high mobility of these lithophile elements is a reason for their increased abundance in the more evolved lavas.

5. Conclusions

An attempt was made through laboratory experiments under room conditions in order to improve the little knowledge so far gained on the thermal parameters of volcanic rocks. It should also be emphasised that these properties are often reported with no information on porosity. This implies that much of the literature data are practically useless (Roy et al., 1981; Rybach, 1988; Zoth and Haenel, 1988). The combined study of index properties and thermal conductivity justifies the anomalous behaviour of rhyolites, which is influenced by porosity and the amount of glass. Samples do not show at the macroscopic scale any anisotropy affecting the thermal conductivity, which in general cannot be considered uniform with depth, as its dependence on temperature as well as pressure must be taken into account. The radioactive heat production varies widely, and the largest values pertain to rhyolitic obsidians. This parameter, which is a scalar and isotropic physical property, is not affected by temperature and pressure, and depends of the geochemical evolution of lavas. The investigated rocks seem in equilibrium in the decay series of the U and Th isotopes.

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