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Internal dose rates of quartz grains separated from fault gouge

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In TL/ESR investigations it is often assumed that quartz is basically free of radioactive elements and that therefore the internal dose rate is negligible. The few measurements that have been rarely carried out, yielded internal dose rates in the range of about 60 to 200 μ Gy/a (e.g. Sutton & Zimmerman 1978, Mejdahl 1987).

In course of an ESR dating study of quartz separated from fault gouge of various sites in Scotland, we have analyzed the U, Th, and K concentrations of the quartz separates as well as the bulk gouge sample by means of neutron activation analysis (NAA).

Samples of the soft clay-like fault gouge were wet sieved through a series of mesh sizes (500 - 63 μm). After storing in 10% HCl for 24h, the samples were washed in de-ionized water and etched with 35% fluorosilicic acid (H₂SiF₆) for 30h to remove feldspars. After washing, the residue was etched with 50% tetrafluoboric acid (HBF₄) at 30 °C for 48h to remove mica (mica-rich samples were 'rolled' in a cellophane film to remove the bulk of such grains). After further washing and air drying the samples were passed through a Franz isodynamic separator at a current of 2 A to separate a dominantly quartz, non-magnetic fraction. If the remaining sample contained notable amounts of pyrite, they were given a 24h bath at 50 °C in conc. nitric acid (HNO₃). The remaining sample was then put in hydrofluoric acid (HF) for 2 h to remove a surface layer of about 20 µm that has been subject to external radiation. A final washing in de-ionized water in a sonic bath for 30 min removed any traces of acid and any particles adhering to the surfaces of the grains. Any remaining non-quartz grains were removed by hand picking under a binocular microscope.

Table 1 gives the sample locations, grain sizes and a short macroscopic and microscopic description of the samples. The macroscopic colours are either associated with mica inclusions or the occurrence of coloured grains, which can be attributed to iron (orange - yellow), copper (green-blue), Fe₂O₃ and Al₂O₃ (purple), Al₂O₃ (dark brown) or Li₂O or MnO (red) in the crystal lattice. White frosty grains are due to a large amount of fluid inclusions in the crystals and HF attack on the grain surfaces. The 'aggregates' are quartz- muscovite intergrowths, a slight yellow coloration can be attributed to pinite. Some samples were X-rayed and the results are also described in table 1.

Table 2 gives the neutron activation results, beta self absorption factors (after Mejdahl 1979) and calculated dose rates using an assumed alpha ef-ficiency of 0.1 and

dose rate conversion factors from Nambi & Aitken (1986). The detection limits of the analyses are 0.1 ppm for U and Th and 0.01% for K. The external beta and gamma dose rates are infinite matrix doses for the respective grain sizes calculated from the NAA results of the bulk gouge samples. The correct value may vary due to radioactive inhomogeneity at the site and variable water contents (mainly for external beta dose rate).

As can be seen from table 2, the concentration of radioactive elements in the quartz separates is rather small compared to that of the respective bulk samples. This leads also to relative small contribution of the internal dose rates to the total dose rate (0.4 to about 7%, in one case 18.9%).

It is remarkable that the separates with low concentrations of U and Th can already be macroscopically recognized. All white samples (except 714B and 717B) have U and Th concentrations below 0.5 ppm. Higher concentrations are connected with colour changes. In the coarser fractions, the internal dose rate of the essentially pure quartz separates does not exceed 3% of the total dose rate. In the finer fractions of these separates, the internal dose rate is lower than 4%. The internal beta dose rate rarely exceeds a third of the internal alpha dose rate and seems therefore negligible although K-concentrations of more than 1% have been measured. Mejdahl (1987) suggested a relationship between U and Th concentrations in quartz. The plot of U- concentration versus Th concentration (without samples 713A and 718B) yields a regression line with Th(ppm) = 1.53 U(ppm) - 0.111. The correlation coefficient is 0.812.

The XRD analysis was not particular conclusive. Sample 718B with the highest concentration of radioactive elements shows besides a quartz spectrum only one additional peak at 3.534. Samples 723B and 725B show some peaks between 2.56 and 4.44, which could not clearly be attributed to common minerals. The other runs yielded basically pure quartz spectra.

In case of the more contaminated separates, the higher concentrations of radioactive elements seem to be linked to the occurrence of non-quartz minerals and hence, the calculated internal dose rate may be an overestimate. The conclusion of the present study is that the internal dose rate is indeed only a minor contribution to the total dose rate. However, the external dose rate is exceptionally high $(4,000 \text{ to } 19,000 \,\mu\text{Gy/a})$, the host rock consists of pegmatites or of Precambrian metamorphic rocks. In many environments such as beach sands or sand dunes, an alpha dose rate of about 120 %Gy/a as generated by

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0.3 ppm U and 0.5 ppm Th does not seem to be negligible. In these cases it also does not seem advisable to calculate the Th concentration from the U- analysis, because this procedure may cause a large uncertainty in the assessment of the internal Th dose rate.

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Table 1. Description of the samples

No.	Site	Grain size (µm)	Colour	Comments				
710	Glen Gloy (W)	250-355	grey	mica inclusions, few red grains; quartz: fractured with frosty surface XRD: nearly pure quartz				
711A	Glen Gloy (E)	250-355	grey	as 710, fewer inclusions; quartz: fractured with frosted surface, some transparent not fractured				
В		125-180	grey	as 711A, some fine grained aggregates				
712A B	Kinloch Houm	250-355 125-180	white brown	nearly pure quartz: extremely fractured, frosty surface, some clear transparent many yellow-red grains; quartz: mostly fractured with frosty surface, some clear transparent. XRD: pure quartz				
713	Glen Elchaig	250-355	grey	few mica inclusions, many yellow or red fractured grains; quartz: slightly fractured frosty surface, more transparent clear grains				
714A	Scardroy 1 (NE)	250-355	brown	Some red grains, mixture of fine grained aggregates and quartz: not strongly fractured, frosty surface. XRD: quantz, small mica peaks.				
В		63-125	white	nearly pure quartz, most frosty surface, many transparent				
715A B	Scardroy 1 (SW)	180-250 63-125	brown-grey brown-grey	some red-yellow grains, quartz: not strongly fractured, frosty surface few yellow grains, quartz: most with frosty surface, some fine grained aggregates, some clear transparent. XRD: pure quartz				
716A	Scardroy 2	250-355	brown-grey	some mica inclusions, rock fragments, quartz: half frosty, half clear transparent, some fine grained aggregates				
В		63-125	grey	yellow red grains; quartz: mixture of frosty and clear transparent grains				
717A	Scardroy 3	250-355	white	nearly pure quartz: not strongly fractured, most with frosty surface, some clear transparent				
В		63-125	white	few yellow red grains, quartz: many fractured with frosty surface, some clear transparent				
718A	Scardroy 4	250-355	grey-red	many yellow red grains, biotite/homblende inclusions; quartz: fractured with frosty surface, few clear				
В		63-125	grey-red	many red yellow grains, rock fragments, biotite/homblende and pyrite inclusions; quartz: heavily fractured with frosty surface, few clear transparent. XR quartz, one unidentified peak				
719A	Scardroy 5a	250-355	white	nearly pure quartz: some heavily fractured frosty, many non-fractured and clear transparent				
В		63-125	white	nearly pure quartz: some heavily fractured, rest in every transition between frosty and clear transparent. XRD: pure quartz				
720A	Scardroy 5b	180-250	grey-brown	few yellow red grains; quartz: not strongly fractured, most with frosty				
В		63-125	white	surface, some clear transparent surface, some fine grained aggregates nearly pure quartz: some heavily fractured, rest in every transition between frosty and clear transparent				
721A .B	Scardroy 5c	250-355 63-125	white white	nearly pure quartz: fractured with frosty surface, few clear transparent nearly pure quartz: some heavily fractured, rest in every transition between frosty and clear transparent				
722A B	Scardroy 5d	250-355 63-125	white white	nearly pure quartz: mixture of fractured frosty and clear non fractured grains nearly pure quartz: some heavily fractured, rest in every transition between frosty and clear transparent				
723A B	Invershiel	250-355 63-125	white brown	nearly pure quartz: mixture of fractured frosty and clear non-fractured grains many yellow red grains; quartz: mostly clear transparent, rest partly fractured with frosty surface. XRD: quartz, some other non-determined peaks				
725A	Coire Eoghainn	250-355	grey brown	many red yellow grains and fine grained aggregates; quartz: fractured				
В	(Glen Cannich)	63-125	brown	with frosty surface, few clear. XRD: pure quartz many yellow red grains and fine grained aggregates; ferric inclusions; quartz: dominantly with frosty surface, many clear transparent XRD: quartz, some other determined peaks				

Table 2: Analytical results and dose rate calculations

Sample			K (%)	absorbed fraction of B dose rate			internal dose rate		external dose rate		internal dose rate
No.	U (ppm)	Th (ppm)		U	Th	K	β (μ(α Gy/a)	β (μGy	/a)	(%)
710Bulk A	36.4 0.6	30.0 1.0	6.4 0.84	0.19	0.25	0.11	99.2	240.8	9625	7255	2.0
711Bulk A B	12.5 0.5 1.3	21.0 0.7 2.0	3.8 0.26 0.99	0.19 0.12	0.25 0.18	0.11 0.05	42.3 73.7	190.8 509.3	4699 5056	3439	2.8 6.4
712Bulk A B	10.7 0.3 0.9	18.0 0.3 1.6	5.5 0.02 0.27	0.19 0.12	0.25 0.18	0.11 0.05	12.3 35.2	105.6 368.5	5651 6067	3491	1.3 4.1
713Bulk A	9.9 0.6	29.0 3.2	4.6 0.35	0.19	0.25	0.11	71.3	403.3	5143	3755	5.1
714Bulk A B	4.7 1.0 1.0	27.0 2.5 0.4	5.0 0.64 0.01	0.19 0.09	0.25 0.14	0.11 0.03	103.4 15.1	462.8 307.7	4771 5251	3157	6.7 3.7
715Bulk A B	2.7 0.5 1.0	4.2 0.6 1.1	5.1 0.15 0.22	0.15 0.09	0.21 0.14	0.08 0.03	24.4 23.1	183.4 359.4	4254 4494	1766	3.3 5.8
716Bulk A B	1.5 0.3 0.6	9.0 0.5 0.6	4.2 0.17 0.07	0.19 0.09	0.25 0.14	0.11 0.03	27.2 12.1	120.4 211.2	3418 3743	1661	2.8 4.0
717Bulk A B	1.0 0.3 1.0	7.4 0.1 0.4	4.5 0.07 0.12	0.19 0.09	0.25 0.14	0.11 0.03	15.3 17.8	90.8 307.7	3541 3873	1594	2.0 5.6
718Bulk A B	3.3 1.2 4.2	21.0 1.0 3.8	4.0 0.04 0.32	0.19 0.09	0.25 0.14	0.11 0.03	44.3 78.8	407.6 1448.8	3749 4125	2442	6.8 18.9
719Bulk A B	1.7 0.1 0.4	12.0 <0.1 0.2	5.2 0.01 0.01	0.19 0.09	0.25 0.14	0.11 0.03	3.7 6.3	27.8 126.0	4232 4634	2083	0.5 1.9
720Bulk A B	3.6 0.3 0.3	9.9 0.5 0.3	5.3 0.20 0.01	0.15 0.09	0.21 0.14	0.08 0.03	22.7 5.4	120.4 105.6	4647 4915	2214	2.0 1.5
721Bulk A B	1.7 0.1 0.3	8.6 <0.1 0.2	5.2 0.02 0.02	0.19 0.09	0.25 0.14	0.11 0.03	4.6 5.3	27.8 98.2	4158 4549	1906	0.5 1.6
722Bulk A B	1.3 0.1 0.4	9.2 0.1 0.3	3.2 0.03 0.04	0.19 0.09	0.25 0.14	0.11 0.03	6.2 7.5	35.2 133.4	2674 2931	1406	1.0 3.1
723Bulk A B	34.5 0.1 1.6	42.0 0.4 3.1	7.0 0.09 1.00	0.19 0.09	0.25 0.14	0.11 0.03	13.8 58.1	57.4 674.1	10094 11192	7811	0.4 3.7
725Bulk A B	5.2 0.4 1.2	28.0 1.1 2.1	5.2 0.17 1.60	0.19 0.09	0.25 0.14	0.11 0.03	34.4 63.5	192.5 488.9	4997 5501	3315	2.7 5.9