Chemical Weathering, First Cycle Quartz Sand, and its bearing on Quartz Arenite

Abhijit Basu

Department of Earth and Atmospheric Sciences, Indiana University, Bloomington, IN 47408, U.S.A. basu@indiana.edu

Abstract

A self-consistent set of experimentally determined rates of mineral dissolution (Francke, 2009) has been used to estimate the relative loss of common constituents of plutonic igneous rocks that supply quartz to sands to be lithified as first-cycle quartz arenite. A first-order decay equation is used setting the decay constant of calcite to one, which renders the time-steps of loss dimensionless but keeps the relative loss of each mineral constant. Calculations show that 99.33% of pure calcite would dissolve only after 5 time-steps. On a relative scale, it would take about 1250, 900, and 1040 time-steps respectively to reduce the original compositions of quartz-bearing mafic plutonic, granodioritic, and granitic rocks to leave >95% undissolved quartz as residue that will qualify as quartz-sand, the precursor to first-cycle quartz arenite. These are very large numbers indicating that chemical dissolution alone, as in chemical weathering by itself, is not sufficient to generate first-cycle quartz sand; accompanied mechanical weathering is necessary. Therefore, it is necessary to re-evaluate many explicitly stated inferences of warm, humid climate in provenance studies of first-cycle quartz arenites.

Introduction

In the context of quartz arenite (Pettijohn, 1957; Dott, 1964; Folk, 1980; Garzanti, 2019a), quartz sand is a body of natural material in which 95% or more of the sand-sized fraction $(0.0625 \text{ mm} - 2 \text{ mm})$ consists of clastic quartz. In the context of first cycle quartz arenite, first cycle quartz sand is a product of weathering of plutonic or medium to high grade metamorphic rocks, i.e., *not* derived from pre-existing sedimentary rocks. Prolonged weathering of very low relief granitic terrains under hot, humid, tropical climate and ample vegetation can eventually lead to a product of quartz and clay (Krynine, 1935; Suttner et al. 1981; Johnsson and Stallard, 1988; Soegaard and Eriksson, 1989; Swain and Gorana, 2009; van de Kamp 2010; Garzanti et al., 2013; Lorentzen et al., 2019; Khan et al., 2020 and references therein). Preferential mechanical disintegration of detrital feldspars and lithic fragments, especially through reworking on a sea beach, may generate a quartz-rich sand fraction depending on the composition of the sand reaching a beach (Sedimentation Seminar (Ferree et al.) 1988). However, authentication of any dramatic petrologic change by comparing the composition of a set of "prebeach and beach sands" are few, if any. Rather, Ruxton (1970) documents a quartz-rich lateritic soil on a seacliff and lithic sand on the beach (see also, Basu, 1985).

First cycle quartz sand upon lithification would produce first cycle quartz arenite. First cycle quartzo-feldspathic sand may also produce "first cycle" quartz arenite if diagenetic processes remove the feldspars (McBride, 1987; Chandler, 1988), replace the feldspars with silica (Wallace, 1976), or alter the feldspars to clay (Sorby, 1859; Brenchley, 1969; Valloni et al., 1991) in which case, however, the arenitic texture moves towards that of wacke (Dott, 1964). Indeed, in its pure form, usually silica cemented and white, first cycle quartz arenite is a fascinating rock that rivals "pure single malt Scotch whiskey" (Dott, 2003). Amajor (1987), however, stresses that "… most ancient quartz arenites are multicycle in origin". We refer the reader to Garzanti et al. (2019) who has discussed the quartz arenite problem with great erudition followed by Lorentzen et al. (2020) with a dissenting opinion.

The qualitative arguments for the production of quartz sand through weathering are so strong that the inverse, i.e., the occurrence of ancient first cycle quartz arenites has been used to infer the paleoclimate – warm and humid – of source regions (e.g., Dutta and Wheat, 1993; Avigad et al. 2005; Chakraborty and Sensarma, 2008; Quasim et al., 2017). Petrographic evidence of recycling such as presence of mudclasts (Choudhuri, et al., 2020), grain rounding (Suttner and Dutta, 1986; Mehring and McBride, 2007), or abraded quartz overgrowths (Ojakanagas, 1963; Johnsson and Stallard, 1988; Critelli et al., 2013; Garzanti et al., 2013; Basu et al, 2013) are not often discussed in these studies on paleoclimate. Short rivers depositing sand in a basin commonly run through rugged topography and often

change drainage patterns thus preventing long-lived weathering and resulting in deposition of arkosic or lithic sands (Krynine, 1935; Stevens Goddard et al., 2020). Long rivers, such as the Amazon, Congo, Ganges, St. Lawrence, etc., capable of temporarily storing sand on floodplains where further chemical weathering takes place, however, do also drain sedimentary rocks such that the river sands become a mixture of first and recycled detritus. These rivers also flow across different climatic zones. So far, no attempt has been made to quantify the extents of these variables in the context of the origin of first cycle quartz sand.

Sand production is a function of chemical, mechanical, and bio-weathering (Pettijohn et al., 1972; Nesbitt et al., 1997; Napieralski et al., 2019; Caracciolo, 2020). This paper addresses only chemical weathering as a complement to primarily mechanical breakdown of clastic grains during fluvial transport and beach reworking in the production of quartz sand (Blatt, 1967; Sedimentation Seminar (Ferree et al.), 1988; and references therein).

The purpose of this investigation is to assess quantitatively the relative survival of detrital minerals through progressive dissolution, and eventual production of quartz sand and quartz arenite. A theoretical approach is taken to assess the relative efficiencies of producing quartz sand from common plutonic rocks. A simple first order decay equation and experimentally determined relative rates of dissolution of common minerals are used to calculate how many steps of decay – from source to sink – would be necessary to convert a plutonic rock to quartz sand. Mechanical strengths of rocks and minerals are not considered.

Assumptions, Parameters, Proxies and Rationale

It is assumed that the loss of detrital minerals progresses exponentially as a first-order process, which can be monitored using the equation $P_t = P_0 e^{-\lambda(m)t}$, where P_0 is the original population of mineral "m", P_t is the population of mineral "m" after time "t" since the beginning of the process of mineral loss, and $\lambda(m)$ is the rate constant of loss of mineral "m". In reality, the absolute amount of loss of mineral "m", however, increases at a greater rate as its grain size decreases with concomitant increase in specific surface area. This increase is countered by the precipitation of the alteration product (e.g., clay) that forms a coat on mineral grains and prevents the solvent (~water) from dissolving the mineral efficiently, and, if the solvent reaches saturation before being "washed away". It is further assumed that the reactive surfaces of all mineral grains are fully available for dissolution throughout the complete process of weathering. In reality, it is not so

Table 1. Mineral Dissolution Rates (~λ values)

Note: Blank in column 3 indicates that the value in column 4 is estimated from other data.

Nomenclature in Francke's Table 2 and Table 3 are not consistent; for example, Ca-Plag (An66) is called anorthite.

because products of dissolution may coat a mineral grain and the solvent may change its composition. Note that the rate of grain coating and the rate of solvent replenishment cannot be quantified. Further, experiments by Holdren and Speyer (1985, 1987) show that grain surface defects are more important than grain surface area in facilitating dissolution reactions. Hence,

the assumption is a pragmatic compromise for the purpose.

Franke's (2009) experimental data on rates of dissolution of minerals have been used as quantitative guides to estimate λ. Although not as comprehensive as the huge compilation in Palandri and Kharaka (2004), this dataset is internally consistent, i.e., experiments were conducted at the same P, T and pH for all 39 minerals in the same laboratory by the same investigator. The experimentally determined rates of dissolution of common rock forming minerals at 20° C and $pH = 5.5$ are given in Table 1. To assess quantitatively the relative loss of minerals through progressive dissolution under principally chemical

weathering, λm has been normalized to the dissolution rate of calcite (cc) such that λ cc = 1 (dimensionless). Francke (2009) was not able to determine the rates of dissolution of quartz (qz) and zircon (zr). Palandri and Kharaka's compilation (2004) indicates that the relative rates of dissolution of quartz and zircon under similar conditions would be a few orders of magnitude lower. Values of 0.0001 have been assigned to λ qz and λ zr. Similar reasoning and extrapolations have been used to assign λm for minerals for which Francke (2009) did not have direct measurements (Table 1).

Because λm is dimensionless, results of calculations represent relative time-steps in the process of chemical dissolution of the minerals but not absolute

time. Each step is an integral of intensity and duration of chemical duration (~ weathering) that is experienced equally by all minerals in an assemblage of interest. It follows that the amount of absolute dissolution suffered

by each mineral in a time-step is proportionate to their respective values of λm. For example, in five time-steps calcite would be reduced from 100% to 0.67%, plagioclase $({\sim}An_{65})$ to 71.64%, and K-feldspar to 97.94%.

Calculations and Results.

For assessing the effects of chemical dissolution of source rocks of sediments, modal compositions of a few common plutonic parent rocks were used as starting material, which include 13 granites, 5 granodiorites, 3 tonalites, 2 diorites, one gabbro, and one adamellite (Table 2). The samples were treated in three groups, i.e., granite-adamellite, granodiorite-tonalite, and diorite-gabbro.

Table 3: Normalized compositions of undissolved residuals from three suites of plutonic rocks after stepwise dissolution **Granite**

Time-Steps	Quartz	K-Felds	Plag	Musc	Biot	Hbl	ClinoPx	OrthoPx	Opaque	Chlorite	Total
$\boldsymbol{0}$	29.92	38.84	19.87	0.12	9.35	0.36	0.22	0.00	0.70	0.62	100
$\mathbf{1}$	30.03	38.82	19.86	0.12	9.36	0.35	0.21	$0.00\,$	0.68	0.57	$100\,$
5	30.44	38.72	19.81	0.12	9.39	0.30	0.20	$0.00\,$	0.61	0.40	100
10	30.94	38.56	19.73	0.12	9.42	0.25	0.19	$0.00\,$	0.54	0.26	100
20	31.89	38.15	19.52	0.13	9.46	0.17	0.16	$0.00\,$	0.41	0.11	100
30	32.80	37.69	19.28	0.13	9.49	0.11	0.14	$0.00\,$	0.31	0.05	100
40	33.71	37.18	19.02	0.13	9.50	0.08	0.12	$0.00\,$	0.24	0.02	100
50	34.61	36.65	18.75	0.14	9.51	0.05	0.10	$0.00\,$	0.18	$0.01\,$	100
60	35.51	36.11	18.47	0.14	9.51	0.04	0.09	$0.00\,$	0.14	0.00	100
70	36.41	35.55	18.19	0.14	9.50	0.02	0.07	$0.00\,$	0.11	$0.00\,$	100
80	37.32	34.99	17.90	0.15	9.49	0.02	0.06	$0.00\,$	0.08	$0.00\,$	100
90	38.23	34.41	17.60	0.15	9.48	0.01	0.05	$0.00\,$	0.06	0.00	100
100	39.15	33.83	17.31	0.15	9.46	$0.01\,$	0.05	0.00	0.05	0.00	100
200	48.53	27.93	14.29	0.18	9.07	0.00	$0.01\,$	$0.00\,$	$0.00\,$	$0.00\,$	100
300	57.89	22.18	11.35	0.20	8.37	0.00	$0.00\,$	$0.00\,$	0.00	$0.00\,$	100
400	66.63	17.00	8.70	0.22	7.45	0.00	0.00	$0.00\,$	0.00	0.00	100
500	74.26	12.62	6.45	0.24	6.43	0.00	0.00	0.00	0.00	$0.00\,$	100
600	80.58	9.12	4.66	0.25	5.40	0.00	0.00	$0.00\,$	0.00	$0.00\,$	100
700	85.57	6.45	3.30	0.25	4.43	0.00	0.00	$0.00\,$	0.00	0.00	100
800	89.39	4.48	2.29	0.25	3.58	0.00	$0.00\,$	$0.00\,$	0.00	$0.00\,$	100
900	92.24	3.08	1.58	0.24	2.86	0.00	0.00	$0.00\,$	0.00	0.00	$100\,$
1000	94.33	2.10	1.07	0.24	2.26	0.00	0.00	0.00	0.00	0.00	100
2000	99.60	0.04	0.02	0.16	0.18	0.00	$0.00\,$	0.00	0.00	$0.00\,$	$100\,$
5000	99.96	0.00	0.00	0.04	0.00	0.00	0.00	0.00	0.00	0.00	100
Granodiorite-Tonalite											
Time-Steps	Quartz	K-Felds	Plag	Musc	Biot	Hbl	ClinoPx	OrthoPx	Opaque	Chlorite	Total
$\boldsymbol{0}$	25.78	10.69	43.41	$0.02\,$	10.67	8.38	0.00	$0.00\,$	0.92	0.14	$100\,$
$\mathbf{1}$	26.02	10.75	43.33	0.02	10.75	8.11	0.00	$0.00\,$	0.90	0.13	100
5	27.00	10.97	42.92	0.02	11.04	7.13	$0.00\,$	$0.00\,$	0.83	0.09	100
10	28.20	11.23	42.31	0.02	11.38	6.05	0.00	$0.00\,$	0.74	0.06	$100\,$
20	30.54	11.67	40.81	0.03	12.01	4.32	0.00	$0.00\,$	0.60	0.03	100
30	32.79	12.03	39.03	0.03	12.57	3.06	$0.00\,$	$0.00\,$	0.47	$0.01\,$	$100\,$
40	34.97	12.32	37.08	0.03	13.07	2.15	0.00	$0.00\,$	0.38	0.01	100
50	37.08	12.55	35.03	0.03	13.51	1.51	0.00	$0.00\,$	0.30	0.00	100
60	39.14	12.71	32.93	0.03	13.89	1.05	0.00	$0.00\,$	0.23	0.00	$100\,$
70	41.15	12.83	30.84	0.03	14.24	0.73	0.00	$0.00\,$	0.18	$0.00\,$	100
80	43.10	12.91	28.78	0.04	14.53	0.50	$0.00\,$	$0.00\,$	0.14	$0.00\,$	100
90	45.01	12.94	26.77	0.04	14.79	0.35	0.00	0.00	0.11	0.00	100
100	46.86	12.94	24.83	0.04	15.01	0.24	$0.00\,$	$0.00\,$	0.08	$0.00\,$	100
200	62.53	11.49	10.42	0.05	15.50	$0.00\,$	$0.00\,$	$0.00\,$	$0.01\,$	$0.00\,$	$100\,$
300	73.14	8.95	3.83	0.06	14.02	0.00	0.00	$0.00\,$	0.00	0.00	100
400	80.19	6.54	1.32	0.06	11.89	0.00	$0.00\,$	0.00	0.00	0.00	100
500	85.11	4.62	0.44	0.06	9.77	0.00	0.00	0.00	0.00	0.00	100

Compositions of plagioclase as counted in petrographic modal analyses were respectively assigned as An_{50} , An_{50-65} , and An_{50} the rates of dissolution of which are available in Franke (2009). Results of step-wise dissolution of the averages of these three groups are given in Table 3. Proportions of undissolved minerals were recalculated to 100% to obtain mineral percentages of the presumed sand fraction after each time-step. Table 4 lists a few vari-

ables in each sample and the averages, which bear upon the discussion of generating first-cycle quartz sand and quartz arenite.

An example of a complete calculation is given in Appendix Table 1 with a granodiorite (CR29; Noyes et al., 1983) as the parent rock with λ_{Pl} = 0.0117. The corresponding Excel File is archived and can also be obtained directly from the author.

Table 4. Rock compositions and time-steps required for 50% loss and attaining 95% quartz

Discussion

Quartz arenite is the extreme and ultimate sandsize product of sedimentary processes (weatheringtransport-deposition-diagenesis/ lithifica-tion) operating on a planetary body. Intensity and duration of weathering, both physical and chemical, from source to sink may produce sands with >95% detrital quartz dissolving or winnowing out other minerals supplied at source. Most beach sands at the mouths of long rivers are extremely quartz-rich (e.g., Potter, 1978; Johnsson et al., 1991; Mehring and McBride, 2007; Garzanti et al., 2015; and references therein). Long rivers commonly derive their detrital load not only from crystalline igneous and medium to high grade metamorphic rocks, but also from sedimentary and low grade metamorphic rocks, i.e., recycled sands (see definition above; also Suttner et al., 1981). If not, i.e., if the detritals are not recycled, the sand is first cycle. If the sand consists of >95% quartz, it would be lithified into a first cycle quartz arenite. Whereas this is intuitive, one may reasonably inquire if we can quantify time-steps of weathering to produce quartz-sand as undissolved residue through mineral dissolution.

Calculations described above show that relative to calcite that dissolves to <50% in less than one timestep, it takes about 23, 125, and 300 time-steps to dissolve 50% of average quartz-bearing mafic plutonic rocks, granodioritic rocks, and granitic rocks respectively (Table 4). Quartz in the remaining 50% undissolved solid residue amounts to 24%, 51%, and 58% respectively. Assuming that the residue remains in the sand-size fraction, it takes about 1250, 900, and 1040 time-steps respectively for the undissolved residue to become first cycle quartz sand (>95% quartz). At those time-steps, nearly 90%, 75%, and 70% of the original parent rocks would also have dissolved. Under normal surface conditions in the tropics of earth, only "prolonged" weathering of low-relief felsic plutonic terrain can generate first-cycle quartz sand (Krynine, 1935; all textbooks since). To do so, weathering must be prolonged by a factor of $10³$ relative to calcite that reduces by ~65% in *one* time-step. Given how lofty high grounds and mountains of limestone, marble, and other carbonate rocks stand today, expecting such "prolonged" weathering is unrealistic.

During weathering, most of the dissolved components precipitate as alteration products such as clay. If we assume that about 10% of the dissolved component stays in solution in river waters, then the ratio of quartz to clay (*sensu latu*) would be approximately 1:8, 1:7, and 1:6 respectively. If quartz and clay were winnowed from each other and deposited as sand and mud respectively, then upon lithification these would be quartz arenite and mudstone respectively. Under such conditions, the proportion of first cycle quartz arenite and mudstone would vary

between 1:6 and 1:8 in a single stratigraphic unit depending upon the assemblage of parent rocks. Thus, everything else remaining the same, every 1 m thickness of quartz arenite in a single stratigraphic succession should be balanced by 6 to 8 m of mudstone. Quartz-free mafic rocks in source areas, such as basalt or andesite in magmatic arcs, will only add to the mudstone budget.

In the above analysis of mineral dissolution,

Figure 1. Evolution of quartz-mafic mineral-feldspar compositions of quartzdiorite-gabbro (diamonds), granodioritic (squares), and granitic (triangles) rocks with progressive dissolution

lithic fragments are not treated as a component in the modal composition of a sandstone. Therefore, the results of intermediate products, i.e., the composition of the sand-product on way to becoming quartz-sand, cannot be interpreted by the standard QFL plots (e.g., Dickinson et al., 1983; Marsaglia, 2004; Devi and Mondal, 2008; Sen et al., 2014; Majumder, 2020; and references therein). Those that consist of primarily sandsize quartz and feldspar without or with very little of other minerals, would be counted as either quartz or feldspar in the commonly used GD method. Mafic minerals (e.g., biotite, amphibole, pyroxene, olivine, Feoxide phases) in the parent rocks would be present in their weathering products albeit in dwindling proportions. The trajectory of the weathering products can be tracked in a Quartz-Mafic Mineral-Feldspar- (QMF) diagram (Fig. 1). Although mafic minerals are primarily sourced from magmatic arcs, the QMF diagram should not be considered analogous to the standard QFL plot. Rather, this diagram may be considered as a limited quantified version of Figure 1 of Blatt (1967; p. 1036) that tracks the "relative effects of sedimentary processes on sand-sized clastic grains".

The diagram illustrates the compositional paths of weathering products of the principal plutonic rocks to become quartz sand. If weathering is stopped at any time-step, the product lithified as a sedimentary rock, and subsequently weathered again, the recycled product will still need many time steps to produce quartz sand. For example, if recycling begins after 100 time-steps then it would still be necessary to go through

about 800 new time-steps to concentrate quartz to >95%. Under conditions in which only 5 time-steps reduces 100% calcite to $\langle 0.7\% \rangle$, 800 is a large number. It is concluded that although possible under extreme conditions, chemical weathering *alone* does not produce first-cycle quartz sand.

Mechanical destruction of feldspars and lithic fragments is necessary to concentrate quartz as residual. In natural stream samples of sand, finer grain size fractions have lesser proportions of feldspars than quartz and very little composite grains, i.e., lithic fragments (e.g., Odom et al., 1976; Basu, 1976; Garzanti, et al., 2009). Transport in a long river and prolonged reworking on a beach can reduce a feldspathic sandy assemblage to quartz sand (Johnsson, 1988; Johnsson et al., 1991). If long rivers from source to sink or if beaches actually remain free of any input of recycled sand from tributaries and aeolian supply in the natural system, will be a matter of discussion in a follow-up note.

There are more than a thousand reports of firstcycle quartz arenites in the literature. All call on primarily chemical and some mechanical weathering for the production of quartz sand. Most explanations include warm and humid climate, low-relief hinterland consisting of felsic plutonic rocks, and long duration weathering. For example, see the discourses by Soegaard and Eriksson (1989) on the Paleoproterozoic Ortega Group in U.S.A.; Chakraborty and Sensarma, (2008) on the Paleoproterozoic Karutola Formation in India; the general discourses by Van de Kamp (2010) and Johnsson et al. (1991); and references therein. If paleotectonic reconstructions indicate rapid erosion and rapid deposition, very toxic atmospheric conditions such as those with high $CO₂$ capable of rapid chemical weathering have been inferred. For example, Avigad et al. (2005) envisage "An unusually corrosive Cambro-Ordovician atmosphere and humid climate enhanced chemical weathering on the vegetation-free landscape" in all Pan-African terrains; Holland et al. (2020) and Jones et al. (2008) postulate acid weathering in greenhouse atmosphere in combination with mechanical weathering to produce the extensive "quartz arenite successions from Paleoproterozoic and Mesoproterozoic strata throughout North America", some of which are adjacent to juvenile arcs.

Because chemical weathering by itself does not produce extensive deposits of quartz sand, inferences

about paleoclimate from the presence of first-cycle quartz arenite in a sedimentary basin are not as robust as some publications may lead us to believe.

Examples cited above and numerous others appear to contradict the inference drawn from the theoretical analysis of experimental data on mineral dissolution. Johnsson and Stallard (1988) contended

that claims to first cycle origin of detrital quartz can only be tested by the "presence of sedimentary lithic fragments and syntaxial quartz overgrowths" as has been demonstrated in lithified quartz arenites, for example by Ojakangas (1963) but not explicitly followed by others. In view of this study, it is advisable to search for abraded overgrowths on detrital quartz grains in the putative first cycle quartz arenites (e.g., Basu et al., 2013; and references therein).

Conclusion

Theoretical consideration of the relative rates of dissolution of common minerals suggests that production of large volumes of first-cycle quartz sand (quartz >95%) from common plutonic rocks on Earth would require unusually long time and possibly unusually toxic atmospheric conditions. Because supply of first-cycle quartz sand is required to produce firstcycle quartz arenites that have been reported from many parts of the earth throughout geological time, new searches for recycled detrital quartz grains are necessary to verify that indeed such first-cycle quartz arenites are free of recycled detrital quartz. Additionally, inferences about paleoclimate on the basis of the presence of quartz arenite appear to require other independent indicators.

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Appendices

Construction of Appendix Table 1

For tracking the progressive relative dissolution of minerals in principal parent rocks through time steps, an Excel Worksheet was set up (for example, see Table 3). Principal minerals that survive as detritals are listed in Column A (Rows 4-14). Appropriate λ values relative to calcite (i.e., λ cc = 1), for each mineral are entered in Column B. Finally, the modal composition (in mineral %) of a parent rock is input in Column C, which is normalized to 100% and appears in Column D. The amount remaining of each mineral after loss in time-steps of 0 to 5000 according to the equation $P_t = P_0 e^{\lambda(m)t}$ (where P_0 is the original population of mineral "m", P_t is the population of mineral "m" after time-step "t" since the beginning of the process of mineral loss, and λm is the rate constant of loss of mineral "m") are in Columns F-AC. These compositions are normalized to 100% and listed in Columns F-AC, Rows 23-33.

Row 18 shows the amount of loss $(P_0 - P_t)$ of the parent rock to dissolution. Row 20 is a nominal check for 100% calcite treated as a parent rock that decreases to ~37% in just one time-step.

In the example (Granodiorite Noyes CR29), 30% of the parent rock is lost after 60 time-steps during which plagioclase (Row 6) decreases from 47.6% (Column D, F) to 23.7% (Column N). And, it would take 800 time-steps to produce quartz-sand (Qz >95%; Column Y, Row 23).

It should be easy for any user of the spreadsheet to input the composition of a suspected parent rock, say R, in Column C to assess possible contribution to a siliciclastic rock from R.

