Oxygen Isotope Thermometry Reveals high magmatic temperatures and short residence times in Yellowstone and other hot–dry rhyolites compared to cold–wet systems
Oxygen isotope thermometry reveals high magmatic temperatures and short residence times in Yellowstone and other hot-dry rhyolites compared to cold-wet systems

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Abstract
The eruption and storage temperatures of rhyolitic magmas are critical factors for understanding the mechanisms of their eruption and petrogenesis. Temperatures are particularly important when comparing the magmatic histories of hot-dry rhyolites from the Yellowstone-Snake River Plain (YSRP) and Iceland to cold-wet rhyolites such as the Bishop Tuff. Here we employ mineral pair oxygen isotope fractionations for estimating rhyolite temperatures independent of pressure and other compositional factors. We report high precision oxygen isotope analyses of quartz, pyroxene, magnetite, and zircon that we use to estimate crystallization and storage temperatures. Temperatures for YSRP and Icelandic rhyolites are highest for quartz-magnetite and quartz-clinopyroxene (~950°C), with lower quartz-zircon (850°C) temperatures that are similar to
estimates of zircon saturation. The magnitude and pattern of these temperatures is consistent with
crystallization from near-liquidus rhyolites. In contrast, oxygen isotope temperatures calculated
for the Bishop and other “cold-wet” type tuffs define low ~760°C temperatures for all three
mineral pairs consistent with prolonged mineral residence at near-solidus conditions.
Preservation of a down-temperature crystallization sequence of hot magnetite and clinopyroxene
with colder zircon in hot-dry YSRP and Icelandic rhyolites suggest <1000 year magma
residence, where magnetite does not have sufficient time to diffusively equilibrate oxygen in a
lower temperature melt. This is consistent with recently determined high precision U-Pb
crystallization ages zircons from the same units indicating magma generation shortly before
eruption.

Keywords: oxygen isotopes, thermometry, magma storage timescales, rhyolites, Yellowstone,
Snake River Plain

Introduction

The origin of rhyolite magmas has been a classic question for igneous petrology, and the
temperature of these magmas is a critical factor in understanding their petrogenesis. Generally
rhyolites can be divided into cold-wet and hot-dry types (e.g., Christiansen 2005). The cold and
wet types contain hydrous minerals, are often crystal-rich, and are typically associated with
convergent margins. Classic examples of this type include the Bishop and similar “monotonous
intermediate” tuffs, interpreted to originate from melt extraction from near-solidus magma
bodies stored as long-lived crystal mushes with >45% crystals (e.g., Bachmann et al. 2002;
Bachmann and Bergantz 2004; Hildreth et al. 2007). On the opposite end of the spectrum are the
hot and dry, plume and rift related-rhyolites, notably represented by the Yellowstone-Snake River Plain (YSRP) of 15 Ma to present (e.g., Branney et al. 2008), and similar aphyric rhyolites of Iceland. These rhyolites typically belong to bimodal basaltic-rhyolitic sequences and have typically low crystallinity (<5-10%) with a paucity of hydrous mineral phases.

A great wealth of geothermometry estimates are available for YSRP rhyolites from mineral-pair analysis (e.g., QUILF, two-pyroxene), zircon saturation, Ti-in-quartz and zircon, and experimental and modeled phase equilibria, and these typically yield high temperatures of up to 950°C (e.g., Cathey et al. 2004, 2009; Vazquez et al. 2009; Watts et al. 2011; Almeev et al. 2012). Many of these geothermometers, however, are highly sensitive to pressure, melt composition, and activities of participating components. Furthermore, recent reformulation of the Zr-saturation thermometer of Watson and Harrison (1983) by Boehnke et al. (2013) yields 30-40°C lower zircon saturation temperatures in many systems. Some phase equilibria studies of Bishop and Yellowstone compositions suggest lower, even <700°C, temperatures (Gardner et al. 2014; Befus et al. 2015; Befus and Gardner in review). Lower temperatures seem more compatible with near-solidus mush storage and extraction models, and several authors now try to apply this model for YSRP centers (e.g., Ellis et al. 2014, Stelten et al. 2015). These very disparate estimates of YSRP rhyolite magma temperatures not only critically influence petrogenetic implications, but also assumed eruption temperatures pertinent to eruption mechanisms and volcanic hazards.

Here we present oxygen isotope thermometry between mineral pairs from YSRP centers as compared to other rhyolites worldwide to provide a unique estimate of rhyolite temperatures (Fig. 1a,b). Stable isotopic fractionations are independent of pressure, melt composition, and phase equilibria, making them particularly valuable as independent tools for determining the
temperature of magmas. Clayton and Epstein (1958) determined that oxygen isotope fractionation between coexisting minerals in igneous rocks could be used to estimate magmatic temperatures. Despite this simple theoretical utility of oxygen isotope thermometers, their use in magmatic systems has been limited. This is due to (1) earlier methods that relied on 10-30 mg of material for analysis and resulted in challenging aggregation of pure mineral separates, especially for crystal-poor YSRP rhyolites; (2) small fractionation factors at high temperatures typical of most, and especially basaltic, igneous rocks coupled with insufficient analytical precision of many earlier $\delta^{18}O$ determinations; and (3) postmagmatic retrogression of mineral-mineral $\Delta^{18}O$ values during slow cooling that limited use especially in plutonic and metamorphic rocks (Eiler et al. 1993). High-precision analyses by laser fluorination of single crystals or small masses of mineral separates (0.7-2.2 mg), however, permits application of this geothermometer for rapidly quenched and lower temperature (700-1000°C) silicic magmas. Application to the highest temperature mafic melts (e.g., 1200°C) is still limited by extremely low fractionation factors, since the high-temperature oxygen isotope fractionation between minerals is proportional to $1/T^2$ (Clayton and Epstein 1958). Bindeman and Valley (2002) demonstrated that oxygen isotope thermometry works for rhyolites and particularly well for rapidly quenched pumices sampling the Bishop magma body, providing new and useful insight into its pre-eruptive conditions.

**Methods**

Laser fluorination oxygen isotope analyses were carried out at the University of Oregon using a 25 W CO$_2$ laser coupled with an MAT 253 10 kV gas source isotope ratio mass spectrometer (IRMS) and purified BrF$_5$ reagent (for further details, see Loewen and Bindeman...
2015). Analyzed mineral separates were pretreated with 48% HF for 3-5 minutes as needed to remove alteration or adhered glass before ~1 mg of material was hand picked for analysis (see electronic supplement for sample weight and raw analytical results). Quartz was selected as single bipyramidal crystals. Clinopyroxenes were selected by color (bright green) and oblique extinction. Magnetite was concentrated with a magnet and hand sorted for freshest grains (black, shiny, and magnetic).

Oxygen isotope fractionation between coexisting solid equilibrium mineral pairs is described by an experimentally determined A-factor and temperature (T, in Kelvin):

\[ T = \frac{\sqrt{A \times 10^6}}{1000 \ln \alpha} \] (1)

where \( 1000 \ln \alpha_{(\text{mineral 1} - \text{mineral 2})} \sim \Delta^{18}O = \delta^{18}O_{\text{mineral 1}} - \delta^{18}O_{\text{mineral 2}} \), for \( \Delta^{18}O < 10 \) (Bottinga and Javoy 1973). Using this relationship, the difference in \( \delta^{18}O \) between coexisting minerals in a volcanic rock can be used to estimate a magmatic temperature. Magnetite, clinopyroxene, and zircon have large A-factors relative to quartz, and, therefore, can be used to calculate a reasonably precise temperature resolution given propagated uncertainty in \( \delta^{18}O \) determinations. We use A-factors determined using a calcite exchange medium from Chiba et al. (1989) for \( \Delta^{18}O_{\text{Qz-Cpx}} \) and \( \Delta^{18}O_{\text{Qz-Mag}} \) of 2.75 and 6.29, respectively. It is important to note that for the purpose of oxygen isotope thermometry in equation 1, clinopyroxene and magnetite are phases of constant composition since cation substitution in these minerals plays an insignificant role in \( ^{18}O-{^{16}}O \) partitioning. The \( \Delta^{18}O_{\text{Qz-Zrn}} \) A-factor is similar to \( \Delta^{18}O_{\text{Qz-Cpx}} \), with a value of 2.64 proposed by Valley et al. (2003) and 2.33 by Trail et al. (2009). Here, we choose to use an
intermediate value of 2.55 corresponding to the observed $\Delta^{18}$O$_{QZ-Zrn}$ and temperature relationship in the Bishop Tuff (Bindeman and Valley 2002).$^1$

As our $2\sigma$ uncertainty in a single mineral $\delta^{18}$O analysis is approximately $\pm0.08\%$ (this study and Loewen and Bindeman 2015), the additive propagated uncertainty ($\sqrt{\sigma_1^2 + \sigma_2^2}$) of any two analyses is $\pm0.12\%$. This will result in an approximately $\pm40^\circ$C in $\Delta^{18}$O$_{QZ-Cpx}$ calculations and $\pm20^\circ$C in $\Delta^{18}$O$_{QZ-Mag}$ calculations. Recycling of antecrystic zircon cores are common in some YSRP units (e.g., Bindeman and Simakin 2014), therefore for units with known antecrystic zircon core compositions we only consider published $\delta^{18}$O zircon rim analyses measured by SIMS. These analyses have higher individual uncertainties of at least $\pm0.2\%$, therefore, these calculated temperatures are $\pm60^\circ$C. We report temperatures calculated by averages of multiple mineral analyses from each sample thus reducing noise from analytical uncertainty.

Since the $\Delta^{18}$O$_{melt-Qz}$ changes very little over expected magmatic temperatures ($<0.2\%$, Fig. 1c), the $\Delta^{18}$O$_{QZ-mineral}$ is proportional to $\Delta^{18}$O$_{melt-mineral}$, with a single offset, where the mineral pairs are clinopyroxene, magnetite, or zircon, all of which are much more sensitive to temperature than quartz. Therefore, our temperature estimates should be accurate as long as the phases crystallized from the same melt composition. Loewen and Bindeman (2015) demonstrated that clinopyroxene in the Yellowstone Central Plateau Member lavas have rhyolitic trace element compositions. We admit that the potential for disequilibrium exists if any mineral phase has an ante- or xenocrystic origin, and for any one sample we cannot verify that

$^1$ Subsequent research has demonstrated that U-Pb dated zircons from the Bishop Tuff are identical to eruption age (Crowley et al. 2007) and thus their $\delta^{18}$O appropriately reflects equilibrium with other phenocryst phases making the Bishop Tuff an ideal case study for testing isotopic fractionations.
the mineral separates analyzed are in equilibrium since mineral separation removes crystals from their petrographic context.

**Results**

New \( \delta^{18}O \) values are reported for clinopyroxene, magnetite, and quartz from the Yellowstone, Heise, Picabo, and Bruneau Jarbidge rhyolites, two new samples from Krafla, Iceland, and compiled data for other YSRP and worldwide units (Table 1; Fig. 1a). For 12 Ma to recent YSRP rhyolites, the median temperature estimated from \( \Delta^{18}O_{Qz-Cpx} \) is 943°C. The highest ~1200°C temperatures recorded from Buffalo Lake and Liddy Hot Springs are beyond any reasonable crystallization temperature for a rhyolite composition, and we suspect these may represent disequilibrium between magmatic \( \delta^{18}O_{Cpx} \) and antecrystic low \( \delta^{18}O_{Qz} \) incorporated during shallow rhyolite remelting. The median 950°C temperature estimated from \( \Delta^{18}O_{Qz-Mag} \) is very similar to that of clinopyroxene, although we note that on a sample-by-sample basis, magnetite temperatures are on average 50°C higher than clinopyroxene. The median temperature estimate from \( \Delta^{18}O_{Qz-Zrn} \) is 847°C and was calculated using the average of previously published SIMS data from zircon rims for each reported sample. On a sample-by-sample basis, zircon temperatures are 70°C lower than clinopyroxene or magnetite temperatures. Average temperatures calculated with \( \Delta^{18}O_{Qz-Zrn} \) are remarkably similar to average whole rock zircon saturation temperatures calculated following Watson and Harrison (1983).

Despite all these individual uncertainties we observe a very consistent overall pattern between minerals. There are no apparent changes in temperature calculated with oxygen isotopes over the 12 Ma YSRP eruptive history that we examine here, and a similar range of temperatures is evident within any one eruptive period as across the entire province (Fig. 1a). In the youngest
Yellowstone volcanism (260 to 79 ka) there is no evidence for decreasing temperatures, as described by Vazquez et al. (2009) using QUILF and Ti-in-quartz thermometry, but there may be a slight increase similar to increases in zircon saturation temperatures (Loewen and Bindeman 2015). Similarly, we do not observe any eastward temperature decrease in YSRP rhyolites as described in Nash et al. (2006).

The YSRP data are directly compared to identically determined oxygen isotope temperatures reported for a number of classic cold-wet tuff deposits, including the Bishop, Cerro Galan, Fish Canyon, lower Bandelier, and Toba tuffs from Bindeman and Valley (2002) and the Kos Plateau (analyses from University of Oregon, reported in Bachmann 2010). Temperatures for mineral pairs from these cold-wet systems are clearly lower and more restricted than YSRP rhyolites (Table 1, Fig. 1b). For the Bishop Tuff, three $\Delta^{18}$O$_{\text{Qz-Cpx}}$ temperatures are 742, 809, and 829°C for the Bishop Tuff. The median temperature from $\Delta^{18}$O$_{\text{Qz-Mag}}$ is 750°C (n=17, range 715-818°C) and is nearly identical to the median $\Delta^{18}$O$_{\text{Qz-Zrn}}$ temperature of 757°C (n=4, range 714-782°C). These lower temperatures are also similar to estimated zircon saturation temperatures (Gualda and Ghiorso 2013). Temperatures from other cold-wet rhyolites follow the same pattern, with nearly identical magnetite and zircon temperatures of ~760°C, closely agreeing with zircon saturation temperature estimates, and classical understanding of eutectic conditions in water-saturated rhyolites. These similar temperatures derived from all minerals in these cold-wet rhyolites contrast the elevated clinopyroxene and magnetite temperatures and relatively lower zircon temperatures in YSRP rhyolites.

Icelandic rhyolites, from another hot-dry system in a rift-type setting have similarly elevated temperatures as the YSRP (Fig. 1a), and especially high magnetite temperatures. In addition to new analyses from Krafla, the average Icelandic values in Table 1 include the 1875
Discussion

Elevated crystallization temperatures

Yellowstone-Snake River Plain oxygen isotope temperatures for clinopyroxene and magnetite reflect the early crystallization of these minerals at elevated liquidus temperatures near 950°C, and clinopyroxene trace element compositions are consistent with origin from very similar rhyolite melts (Loewen and Bindeman 2015). The average clinopyroxene and magnetite temperatures are remarkably consistent with experimentally determined phase equilibria temperatures in the Bruneau Jarbidge rhyolite from Almeev et al. (2012). These experiments determined that the first minerals crystallizing on the liquidus were magnetite followed by clinopyroxene, in agreement with our slightly higher $\Delta^{18}O_{Qz-Mag}$ temperatures. Lower $\delta^{18}O$ temperatures calculated for zircon are consistent with estimates of zircon saturation calculated from either Watson and Harrison (1983) or Boehnke et al. (2013) formulations, but closer to the former, based on either glass or whole rock Zr concentrations. These temperatures are broadly similar to estimates from mineral-pair QUILF calculations although we do not observe similar temperature decreases through time (e.g., Cathey and Nash 2004, 2009; Nash et al. 2006; Vazquez et al. 2009).

In addition, our temperature estimates for the YSRP are remarkably similar to other hot-spot/rift related rhyolites from Iceland (Fig. 1a). Only Lava Creek Tuff member A and the lower
Arbon Valley Tuff have magnetite temperature <900°C, with 710-750°C more in-line with cold and wet rhyolites (Table 1). These two samples are both from early products of each of their eruptive sequences and are some of the only YSRP rhyolites with hydrous minerals (amphibole and biotite, respectively).

The high temperatures cannot be the result of secondary alteration or diffusive retrogression of mineral phases (e.g., Eiler et al. 1993). Zircon and clinopyroxene are especially resistant to low temperature alteration due to extremely small diffusion coefficients (<1x10^{-23} m²/s) in wet or dry conditions at <700°C (Farver 1989; Watson and Cherniak 1997). Even for a small 50 μm zircon crystal, it would take ~3 Ma to equilibrate with surroundings at 700°C in wet conditions. Concern of the possibility for alteration and retrogression of magnetite oxygen isotope values is more justified as diffusion coefficients are 2-5 orders of magnitude higher in magnetite (e.g., Farver 2010). With respect to secondary alteration, the Δ¹⁸O_{Mag-water} is -5 to -7‰ at subsolidus temperatures so water-rock reactions with isotopically light precipitation will only decrease the δ¹⁸O value of magnetite and result in a larger Δ¹⁸O_{Qz-Mag} and therefore lower apparent temperature estimate (Cole et al., 2004). The high temperatures calculated from our magnetite analyses thus can only be achieved by and preserved from high temperature equilibrium crystallization, unless mineral pairs crystallized from melts of different composition. As we have discussed previously, the average temperatures for each mineral that we observe across the YSRP are most likely the result of crystallization temperatures in-line with experimental phase equilibria (Almeev et al. 2012).
Timescales of crystal residence in magmas

Preservation of high oxygen isotope temperatures in mineral phases constrains the time of magma residence after crystallization. The extremely slow oxygen diffusion in zircon and pyroxene means that the oxygen isotopic composition of these phases preserves their crystallization temperature for \(>10^7\) years even at magmatic temperatures (for review, see Farver 2010). Oxygen diffusion is much faster in magnetite with diffusivities 3-4 orders of magnitude faster (Farver 2010). The high observed temperatures calculated from our magnetite analyses could only result from initial crystallization at high temperature as even partial oxygen isotope reequilibration of their rims during crystal residence in a lower temperature magma will quickly increase \(\Delta^{18}O_{QZ-Mag}\) and thus lower calculated temperature.

We can constrain the maximum residence time of magnetite at 850°C that will equilibrate a rim large enough to bring the bulk crystal oxygen isotope value down to but not below the observed 950°C average temperature. Using the experimentally determined diffusion coefficients of Giletti and Hess (1988) for wet conditions and the dry empirically determined coefficients of Sharp (1991), we estimate that the crystal residence time must be \(<100\) years and \(<10,000\) years, respectively, for magnetite up to 400 \(\mu\)m-diameter magnetite crystallizing 40 \(\mu\)m-thick rims. Diffusivities for water-bearing but not water-saturated YSRP magmas are likely between those two endembers, and therefore we consider \(<1000\) years a reasonable estimate for maximum magnetite residence.

High magnetite and clinopyroxene lower zircon temperatures, therefore, reflect two distinctive thermal conditions for YSRP rhyolite magmas: (1) near-liquidus temperatures for clinopyroxene and magnetite crystallization and (2) subsequent short \(<1000\) year residence at \(~100°C\) lower storage at zircon saturation. The pattern of oxygen isotope temperatures from the...
cold-wet systems starkly contrast with our results from the YSRP (Fig. 1). As with YSRP samples, ~810°C temperatures calculated for pyroxene probably reflect crystallization of this phase about 50°C above zircon saturation, and preservation of its oxygen isotope composition due to very slow diffusion rates. Magnetite temperature estimates, however, are identical to zircon temperatures. A simple explanation is that magnetite has equilibrated with the cool and long-lived magma body proposed for these classic cold and wet rhyolites (e.g., Hildreth and Wilson 2007), although magnetite may have also saturated at lower temperatures in these systems.

Implications

Oxygen isotope thermometry strongly supports YSRP and Icelandic rhyolite crystallization from high liquidus temperatures with short (several thousand years or less) residence at lower, zircon-saturated temperatures, following a crystallization sequence in-line with Almeev et al. (2012) phase equilibria experiments. This observation is incompatible with rhyolite extraction from long-lived (10^5-10^6 years) crystal mushes that reside below their zircon saturation temperature. Instead, the most simple explanation for YSRP rhyolite petrogenesis is that they form from rapid, high temperature assembly of partial or wholesale melting of upper crustal rocks generating a hot, near-liquidus melt (e.g., Bindeman and Simakin 2014; Loewen and Bindeman 2015). This melt first crystallized high temperature phases such as magnetite and clinopyroxene before cooling ~100°C and reaching zircon saturation. Storage at this lower temperature is long enough to crystallize eruption-age zircon populations (e.g., Wotzlaw et al. 2014, 2015) but short enough to prevent diffusive reequilibration of magnetite to lower temperature oxygen isotope values. Our results contradict potential extraction of such rhyolites.
from any cold crystal mush achieved through purely mechanical separation without reheating (e.g., Bachmann et al. 2004; Pistone et al. 2015; Stelten et al. 2015). In contrast, cold-wet rhyolites display a limited range of lower oxygen isotope temperatures for all minerals. Similarity of zircon and magnetite is entirely consistent with long residence in a cool, crystal-rich, and more hydrous magma and application of the mush extraction models are more appropriate explanations for their eruption. Our comparison of major silicic systems suggests that aggregate patterns of temperatures calculated for different minerals can be useful in differentiating pre-eruptive storage conditions and generation of rhyolite melts.

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**References**


Figure 1. (a) Oxygen isotope mineral pair temperature calculations for hot-dry Yellowstone-Snake River Plain (YSRP) and Icelandic rhyolites, and (b) the Bishop, Fish Canyon, Toba, Cerro Galan, lower Bandelier, and Kos Plateau Tuffs, shown along-side zircon saturation temperatures (references for compiled data in Table 1). (c) Calculated mineral-melt oxygen isotope fractionation curves at different temperatures, with peak probability values plotted from kernel density estimates of each mineral from hot-dry and cold-wet rhyolites. In hot-dry YSRP and Iceland rhyolites, temperatures calculated with magnetite and clinopyroxene are generally very similar, with the median quartz-magnetite 50°C higher than quartz-clinopyroxene where both minerals are analyzed in the same sample. The median quartz-zircon is 70°C lower than quartz-clinopyroxene. In cold-wet rhyolites, temperatures for all three minerals are lower and remarkably similar. The average quartz-clinopyroxene is ~50°C higher (~810°C) than quartz-magnetite and quartz-zircon (~760°C). Again, zircon saturation temperatures are very similar, and almost 100°C lower than in hot-dry rhyolites. We note that two YSRP deposits, the Lava Creek Tuff member A (LCT-A) and the lower Arbon Valley Tuff (AVT), return similar temperatures to the cold-wet type rhyolites. These two units are the only YSRP units that we have analyzed with hydrous minerals, biotite (bio) and hornblende (hbl), and are the earliest deposits from their respective eruptive sequences.
Table 1. Oxygen isotope thermometry results for Yellowstone-Snake River Plain rhyolites compared to rhyolites worldwide.

<table>
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<th>Group</th>
<th>Unit</th>
<th>Sample</th>
<th>Age (Ma)</th>
<th>Cpx</th>
<th>Mag</th>
<th>Zrn</th>
<th>Qz</th>
<th>Temperature (°C)</th>
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434 * Zircon saturation following Watson and Harrison (1983).  7 Data from Watts et al. (2012).  5 Data from Loewen and Bindeman (2015).  7 Data from Bindeman et al. (2008).  7 Data from Wotzlaw et al. (2015).  7 Data from Watts et al. (2011).  7 Data from Drew et al. (2015).  7 Data from Drew et al. (2013).  7 Data from Cathey and Nash (2004).  7 Data from Bindeman and Simakin (2014).  7 Data are averages from Bindeman and Valley (2002).  7 Data from Gualda et al. (2013).  7 Data from Bachmann et al. (2002).  7 Data from Bindeman (2003).  7 Kos plateau data from Bachmann (2010).  7 Iceland data are from Carley et al. (2011) and Bindeman et al. (2012) for Orpejaukull, Askja, Hekla, Torfajokull, and Austurhorn; two new values for cpx are reported for Krafla; Average isotope ratios are not shown for Iceland due to large differences between eruptive centers.  7 Italic LCT-A analysis is hornblende instead of cpx, Kos Plateau analysis is biotite instead of cpx.
a. hot-dry

b. cold-wet

c. disequilibrium?

disequilibrium?

early deposits

Yellowstone - Snake River Plain

Iceland

Bishop

Fish Canyon

Toba

Cerro Galan

Bandelier

Kos Plateau

hbl

disequilibrium?

early deposits

quartz-clinopyroxene

quartz-magnetite

quartz-zircon

zircon saturation

Quartz-clinopyroxene

Quartz-magnetite

Quartz-zircon

Zircon saturation