Non-centrosymmetry Tests for Potassium Feldspars: An Application of the Second-Harmonic Generator Technique

BY J. A. ZILCZER
Department of Chemistry and Geology, George Mason University, Fairfax, Virginia 22030, USA

AND G. M. LOIAcono
Philips Laboratories, Briarcliff Manor, New York 10510, USA

(Received 22 February 1982; accepted 27 April 1982)

Abstract
Second-harmonic generator tests for non-centrosymmetry have been applied to five monoclinic potassium feldspars. Four samples yielded null results; one specimen displayed a positive test for non-centrosymmetry.

The second-harmonic generator (SHG) test, developed by Kurtz & Perry (1968), and perfected by Dougherty & Kurtz (1976), is an extremely reliable and sensitive detector of non-centrosymmetry in crystals. Several researchers have described the technique itself, and have documented its advantages over previous physical and statistical tests for non-centrosymmetry and acentricity (Woolfson, 1970; Abrahams, 1972; Kurtz, 1972; Dougherty & Kurtz, 1976; Kurtz & Dougherty, 1978). It is the purpose of the current paper to report the results of the first application of second-harmonic analysis (SHA) to feldspar mineralogy.

The natural feldspars examined were selected from the mineral collections of the Department of Mineral Sciences, National Museum of Natural History, Smithsonian Institution, Washington, DC, USA and the Department of Geology, Lafayette College, Easton, Pennsylvania, USA. The samples were transparent and appeared homogeneous in the hand specimen. Sample SI-B18938 was very pale yellow in color. All other specimens were colorless. Electron microprobe, X-ray diffraction and petrographic and electron microscopic analyses, completed in the course of other studies (Zilczer, 1981), confirmed the homogeneity and purity of the samples. All the feldspars were monoclinic and highly potassic (Table 1). Degrees of solid solution were minimal for natural materials. No evidence of exsolution, inclusions, intergrowths or other impurities was found.

To prepare the samples for second-harmonic analysis, single crystals of each specimen were first washed to free them of any possible surface contamination. Crushing in a mullite mortar and sieving through two meshes (numbers 140 and 200) yielded powders of particle size 75 to 105 μm. A second set of powders with particle size <40 μm was also prepared. During these procedures, the samples were kept covered to avoid chance contamination. Microscopic examination of the final powders verified their purity.

The SHG tests were performed at 297 K with a second-harmonic analyzer identical to that described by Dougherty & Kurtz (1976). This instrument uses a neodymium-glass laser with a 1 J output and an optical wavelength of 1.06 μm. To achieve approx-

Table 1. Sample identification and chemical composition

<table>
<thead>
<tr>
<th>Component*</th>
<th>SI-R2824</th>
<th>SI-B21415</th>
<th>SI-B18862</th>
<th>SI-B18938</th>
<th>LC-5491</th>
</tr>
</thead>
<tbody>
<tr>
<td>KAlSi3O8</td>
<td>82.3</td>
<td>86.2</td>
<td>88.2</td>
<td>92.9</td>
<td>96.3</td>
</tr>
<tr>
<td>KFeSi2O8</td>
<td>0.7</td>
<td>-</td>
<td>-</td>
<td>2.1</td>
<td>-</td>
</tr>
<tr>
<td>NaAlSi3O8</td>
<td>15.4</td>
<td>12.0</td>
<td>10.9</td>
<td>4.9</td>
<td>3.7</td>
</tr>
<tr>
<td>NaFeSi2O8</td>
<td>0.1</td>
<td>-</td>
<td>-</td>
<td>0.1</td>
<td>-</td>
</tr>
<tr>
<td>BaAl2Si2O8</td>
<td>1.5</td>
<td>1.7</td>
<td>0.9</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Total</td>
<td>100.0</td>
<td>99.9</td>
<td>100.0</td>
<td>100.0</td>
<td>100.0</td>
</tr>
</tbody>
</table>

*Electron microprobe analyses have been converted to molecular percentages of end-member feldspars (Zilczer, 1981).
†Lafayette College and the Smithsonian Institution have been abbreviated LC and SI, respectively. Sample descriptions employed by these institutions are as follows: SI-R2824: orthoclase, Germany; SI-B21415: sanidine, unknown locality; SI-B18862: adularia, Austria; SI-B18938: orthoclase, Madagascar; LC-5491: adularia, Tavetsch, Switzerland.

0021-8898/82/050540-02$01.00 © 1982 International Union of Crystallography
imate index matching, the samples were placed in oils of refractive index, $n = 1.52$, an average index for these varieties of feldspar. (Small index mismatches of the order of $+0.01$ are unavoidable due to birefringence.) Parallel analyses conducted for both the coarser and the finer powders yielded the same results. Four of the five specimens did not generate a second harmonic when examined down to a signal level of approximately $10^{-2}$ of $\alpha$-quartz. The Tavetsch adularia (LC-5491) gave a positive response and exhibited a second-harmonic intensity of the order of one-thirtieth ($1/30$) of the quartz standard.

In evaluating these tests, the results for the first four samples must be considered null. These specimens still could be non-centrosymmetric, but at a level detectable only with sensitivities beyond $10^{-2}$ of quartz. Sample LC-5491 was the one specimen which displayed a positive test for non-centrosymmetry. It may be noted that this sample was the most 'classical' adularia specimen in the sample suite. Its chemistry was closest to pure $\text{KAISi}_3\text{O}_8$. Its morphology adhered most closely to that typical of adularias (i.e. dominant development of the $\{110\}$ form).

These second-harmonic analyses tend to support the original findings of Hankel (1877) and Taylor (1933). Hankel reported pyroelectric responses for several of the feldspars he tested. When Taylor solved the first monoclinic feldspar structure he contended that its 'real' symmetry must be non-centrosymmetric. The suggestion that, in at least some feldspars, centrosymmetry may be approximate rather than rigorous, would not materially alter the basic results of average structure determinations.* However, as modern studies proceed, SHG tests and their results should prove to be useful auxiliary tools for evaluating the detailed structures of feldspars and other minerals.

Portions of this research were supported by a Smithsonian Predoctoral Fellowship awarded to J. A. Zilczer (1979–81).

*Average feldspar structures have traditionally been refined in holohedral space groups. In one notable exception, Toman & Frueh (1973) reported statistically significant improvement for an intermediate plagioclase refined in space group $C1$ rather than $C1$.

**References**


