

## Lu<sub>2</sub>SiO<sub>5</sub> by single-crystal X-ray and neutron diffraction

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The structure of dilutetium silicon pentaoxide, Lu<sub>2</sub>SiO<sub>5</sub>, has isolated ionic SiO<sub>4</sub> tetrahedral units and non-Si-bonded O atoms in distorted OLu<sub>4</sub> tetrahedra. The OLu<sub>4</sub> tetrahedra form edge-sharing infinite chains and double O<sub>2</sub>Lu<sub>6</sub> tetrahedra along the *c* axis. The edge-sharing chains are connected to the O<sub>2</sub>Lu<sub>6</sub> double tetrahedra by isolated SiO<sub>4</sub> units. The structure has been determined by neutron diffraction.

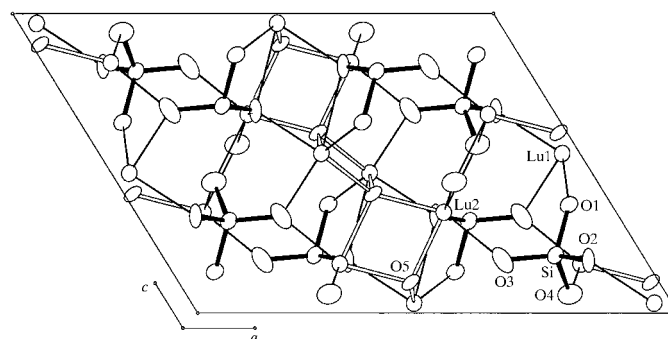
### Comment

Of all known scintillators, cerium-doped Lu<sub>2</sub>SiO<sub>5</sub> (LSO) has the best combination of density, atomic number, light output and speed for the detection of 511 keV annihilation photons in positron emission tomography (PET). PET is a medical imaging technique capable of measuring the concentration of labelled compounds in the human body as a function of time; it is an efficient method for measuring regional biochemical and physiological functions. A large number of these dynamic tracer studies have been conducted throughout the world, resulting in important discoveries in heart disease, brain disorders and cancer.

Since 1980, the scintillation crystal Bi<sub>4</sub>Ge<sub>3</sub>O<sub>12</sub> (BGO) has been used for the detection of the 511 keV annihilation photons in PET, primarily because its density and atomic number give it a higher photoelectric efficiency than more conventional scintillators, such as NaI:Tl. More recently, the scintillation crystal Lu<sub>2</sub>SiO<sub>5</sub>:Ce (LSO; Melcher & Schweitzer, 1992) was found to have a similar detection efficiency to BGO whilst being seven times faster and having a four times higher light output. This crystal is now being grown in large quantities by CTI Positron Systems Inc., Knoxville, Tennessee, USA, one of the largest producers of positron tomographs worldwide. This corporation has now constructed the first LSO positron tomograph, which contains over 120 000 2.1 × 2.1 × 7.5 mm LSO crystals. It is anticipated that the use of LSO in medical imaging will increase steadily in the foreseeable future.

It is necessary to understand the underlying scintillation mechanisms in order to develop even better scintillator materials for use in medical imaging, nuclear physics, high-energy physics and astrophysics. This is being performed mainly through electronic structure calculations, *i.e.* embedded cluster calculations in a Hartree–Fock approach or periodic band structure calculations using a full-potential linear muffin tin orbital approach. Structural information on the title material is therefore essential.

A single-crystal X-ray data set was collected on a Siemens CCD diffractometer and the structure was solved using direct methods. Because of the poor accuracy achieved for the positional parameters of the O atoms, it was also decided to collect a single-crystal neutron diffraction data set. This was performed at the medium-flux R2 steady-state reactor at Studsvik, Sweden.



**Figure 1**

The structure of LSO, with displacement ellipsoids at the 50% probability level. Si–O bonds are filled, bonds in the OLu<sub>4</sub> tetrahedra are open and other Lu–O bonds are shown as thin lines.

The structure of Lu<sub>2</sub>SiO<sub>5</sub> is shown in Fig. 1. Infinite chains of edge-sharing OLu<sub>4</sub> tetrahedra run along the *c* axis. These are connected by SiO<sub>4</sub> tetrahedra. The structure can thus be described as SiO<sub>4</sub> tetrahedra and non-Si-bonded O atoms surrounded by four Lu atoms in a distorted tetrahedron. This is the well known structure for the smaller rare-earth (Dy–Lu) orthosilicates (Felsche, 1971, 1973; Anan'eva *et al.*, 1981).

### Experimental

LSO is commercially available from CTI Positron Systems Inc., Knoxville, Tennessee, USA, and crystals were grown by the Czochralski method (Pamplin, 1980).

#### Crystal data

Lu<sub>2</sub>SiO<sub>5</sub>  
*M<sub>r</sub>* = 458.02  
 Monoclinic, *C*2/*c*  
*a* = 14.2774 (7) Å  
*b* = 6.6398 (4) Å  
*c* = 10.2465 (6) Å  
 $\beta$  = 122.224 (1)°  
*V* = 821.74 (8) Å<sup>3</sup>  
*Z* = 8  
*D<sub>x</sub>* = 7.36 Mg m<sup>-3</sup>  
 Cell parameters determined using Mo *K*α radiation

Structure determined using reactor neutron radiation  
 $\lambda$  = 1.20700 Å  
 Cell parameters from 1391 reflections  
 $\theta$  = 1.5–22.5°  
 $\mu$  = 0.16 mm<sup>-1</sup>  
*T* = 293 K  
 Prismatic, colourless  
 5.6 × 2.8 × 2.8 mm

## Data collection

ARACOR diffractometer	$R_{\text{int}} = 0.047$
$\theta/2\theta$ scans	$\theta_{\text{max}} = 52^\circ$
Absorption correction: by integration (ABSSTOE; Lundgren, 1983)	$h = -18 \rightarrow 0$
$T_{\text{min}} = 0.607$ , $T_{\text{max}} = 0.684$	$k = 0 \rightarrow 8$
999 measured reflections	$l = -11 \rightarrow 13$
961 independent reflections	3 standard reflections
949 reflections with $I_{\text{net}} > -10\sigma(I_{\text{net}})$	every 33 reflections
	intensity decay: 0.4%

## Refinement

Refinement on $F^2$	$(\Delta/\sigma)_{\text{max}} = 0.050$
$R(F) = 0.035$	$\Delta\rho_{\text{max}} = 0.09 \text{ barn } \text{\AA}^{-3}$
$wR(F^2) = 0.078$	$\Delta\rho_{\text{min}} = -0.06 \text{ barn } \text{\AA}^{-3}$
$S = 2.91$	Extinction correction: B-C type 1
949 reflections	Gaussian anisotropic
79 parameters	Extinction coefficient: see below
Weighting scheme: $1/\sigma^2(F)$ modified for experimental instability	

Table 1

Selected bond lengths ( $\text{\AA}$ ).

Lu1—O1	2.2561 (10)	Lu2 <sup>v</sup> —O4 <sup>v</sup>	2.2378 (10)
Lu1—O1 <sup>i</sup>	2.2949 (10)	Lu2 <sup>v</sup> —O4 <sup>viii</sup>	2.2356 (11)
Lu1—O2	2.6163 (11)	Lu2 <sup>v</sup> —O5 <sup>v</sup>	2.1652 (10)
Lu1—O2 <sup>ii</sup>	2.3301 (10)	Lu2 <sup>v</sup> —O5 <sup>ix</sup>	2.2642 (10)
Lu1—O3 <sup>iii</sup>	2.2756 (10)	Si—O1	1.6242 (13)
Lu1—O5 <sup>iv</sup>	2.1598 (10)	Si—O2	1.6395 (13)
Lu1—O5 <sup>vii</sup>	2.3432 (10)	Si—O3	1.6138 (12)
Lu2 <sup>v</sup> —O2 <sup>vi</sup>	2.2346 (10)	Si—O4	1.6214 (13)
Lu2 <sup>v</sup> —O3 <sup>viii</sup>	2.2350 (10)		

Symmetry codes: (i)  $1-x, 1-y, 1-z$ ; (ii)  $1-x, y, \frac{1}{2}-z$ ; (iii)  $\frac{1}{2}+x, \frac{3}{2}-y, \frac{1}{2}+z$ ; (iv)  $\frac{1}{2}-x, \frac{1}{2}+y, \frac{1}{2}-z$ ; (v)  $x, y, 1+z$ ; (vi)  $\frac{1}{2}-x, \frac{3}{2}-y, 1-z$ ; (vii)  $x, 1-y, \frac{1}{2}+z$ ; (viii)  $\frac{1}{2}-x, \frac{1}{2}-y, 1-z$ ; (ix)  $-x, y, \frac{1}{2}-z$ .

The lattice parameters, space-group symmetry and initial coordinates were determined from a single-crystal X-ray study. Because of high X-ray absorption, refinement was carried out with intensities measured by neutron diffraction (only the data from the neutron diffraction are archived with the CIF). A crystal of  $\text{Lu}_2\text{SiO}_5$  (obtained from CTI Positron Systems Inc.) was mounted on an aluminium pin for the data collection. Net intensities were extracted from step-scanned profiles using the algorithm of Lehman & Larsen (1974). Standard reflection intensities were analyzed (McCandlish *et al.*, 1975) and all intensities and their standard uncertainties were modified according to the variation of the standard reflections, which could be caused by crystal decomposition or instrumental instability. These modified standard uncertainties were later used, unaltered, as weights in the least-squares refinement. After absorption and Lorentz corrections, the atomic parameters from the X-ray structure determination were used as starting parameters for a refinement of the structure based on the neutron diffraction data. An anisotropic

extinction correction was applied [extinction coefficients  $2.06 (9) \times 10^9$ ,  $1.20 (9) \times 10^9$ ,  $1.71 (10) \times 10^9$ ,  $8.3 (8) \times 10^8$ ,  $-1.34 (7) \times 10^9$  and  $-5.3 (7) \times 10^8$ ], but despite this, 50 of the most extinction-affected reflections had to be omitted from the refinement. The largest peak in the final residual scattering power density was found close to atom O3.

Data collection: CRYO (ARACOR, 1981); cell refinement: SAINT (Siemens, 1995); data reduction: ARACOR (Lundgren, 1983); program(s) used to solve structure: SIR92 (Altomare *et al.*, 1993); program(s) used to refine structure: DUPALS (Lundgren, 1983); molecular graphics: ORTEPII (Johnson, 1976); software used to prepare material for publication: DISTAN (Lundgren, 1983).

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Supplementary data for this paper are available from the IUCr electronic archives (Reference: BR1287). Services for accessing these data are described at the back of the journal.

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