^{151}Eu Mössbauer Spectroscopic Studies of "After Effects" of the Electron Capture Decay of ^{151}Gd

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Summary Mössbauer spectroscopy has been used to observe the production of an 'anomalous' charge state, $^{151}Eu^{2+}$, following the electron capture decay of $^{151}Gd^{3+}$ incorporated in $Er_2(C_2O_4)_3,10H_2O$.

MÖSSBAUER studies of 'anomalous' charge states produced by radioactive decay of lanthanide atoms are of particular interest because the chemical effects associated with a change in atomic number are minimised when the transition occurs between adjacent members of the lanthanide series. Relatively few Mössbauer studies of "after effects" have been reported for lanthanide elements. Probst[1] has reported observing $^{151}Eu^{2+}$ produced by beta-decay of $^{151}Sm^{3+}$ incorporated in anhydrous $LaCl_3$. Khurgin, Ofer, and Rakavy[2] have observed the presence of both $^{161}Dy^{4+}$ and $^{161}Dy^{3+}$ produced by beta-decay of ^{161}Tb incorporated in CeO_2. We report the first experimental evidence for the production of $^{151}Eu^{2+}$ following the electron capture decay of $^{151}Gd^{3+}$ incorporated in $Er_2(C_2O_4)_3,10H_2O$.

1 mCi of ^{151}Gd was prepared by irradiating a $^{151}Eu_2O_3$ target with 16 MeV deuterons; the ^{151}Gd was isolated from the target by solvent extraction and purified by ion-exchange.[3] The carrier-free ^{151}Gd was co-precipitated with erbium and incorporated, in turn, into the following compounds, which were used as Mössbauer sources: $^{151}Gd_2O_3$ (in Er_2O_3), $^{151}GdF_3,2H_2O$ (in $ErF_3,2H_2O$), and $^{151}Gd_2(C_2O_4)_3,10H_2O$ [in $Er_2(C_2O_4)_3,10H_2O$]. The ^{151}Eu Mössbauer spectra of the three source compounds were determined at room temperature and 80 K against an Eu_2O_3 absorber, isotopically enriched in ^{151}Eu, maintained at room temperature.

The oxide and fluoride sources, at room temperature and 80 K, exhibited only a single resonance line characteristic of the $^{151}Eu^{3+}$ charge state. However, two resonances were observed in the spectra of the ^{151}Gd doped $Er_2(C_2O_4)_3,10H_2O$

source at both room temperature and 80 K, the intensities of both resonances being greater at 80 K than at room temperature. The Figure shows the 80 K spectrum of a ^{151}Gd doped $Er_2(C_2O_4)_3,10H_2O$ source; the resonance at $-0{\cdot}31 \pm 0{\cdot}10$ mm s^{-1}, width 4·3 mm s^{-1}, is characteristic of the $^{151}Eu^{3+}$ charge state, and the resonance at $+12{\cdot}6 \pm 0{\cdot}2$ mm s^{-1} is characteristic of $^{151}Eu^{2+}$ in the source. The isomer shifts are given relative to a $^{151}GdF_3,2H_2O$ source at room temperature. In order to characterise the resonance at $12{\cdot}6 \pm 0{\cdot}2$ mm s^{-1}, europous oxalate monohydrate, $Eu(C_2O_4),H_2O$, was prepared and its absorption spectrum determined at 80 K with a $^{151}Gd_2O_3$ source. The $^{151}Eu^{2+}$ resonance had an isomer shift of $-12{\cdot}6 \pm 0{\cdot}1$ mm s^{-1} (relative to $EuF_3,2H_2O$) and a width of 5·2 mm s^{-1}. Since the sign of the isomer shift is changed when the roles of source and absorber are transposed these observations provide good evidence that the $^{151}Eu^{2+}$ species produced by electron capture decay are in a europous oxalate environment.

The fact that a $^{151}Eu^{2+}$ resonance was observed with the oxalate and not with the oxide or fluoride may be explained in terms of a radiolytic mechanism following the primary event. Since the oxalate ion is known to exhibit reducing properties when subjected to radiolysis,[4,5] it is probable that in a significant fraction of the events, the Auger cascade produces, by radiolysis, a sufficient number of reducing species in the immediate vicinity of the ^{151}Eu daughter atom, to reduce it to the bivalent state prior to the emission of the 21·7 keV Mössbauer γ-ray. This means that the radiolytic mechanism must be complete in a time less than 10^{-9} s. The width of the $^{151}Eu^{2+}$ resonance produced by the electron capture decay of ^{151}Gd is considerably greater than that of the $^{151}Eu^{2+}$ resonance in the spectrum of $Eu(C_2O_4),H_2O$, indicating that the radiolytic process has damaged the lattice around the decayed atoms.

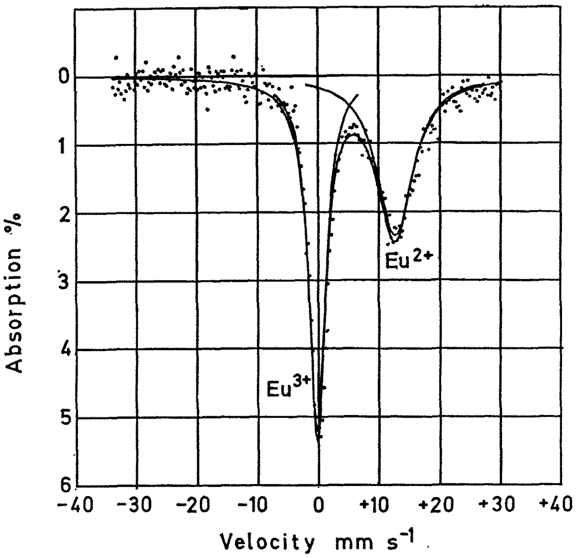

FIGURE. *Mössbauer spectrum, at* 80 K, *of a* ^{151}Gd *doped* $Er_2(C_2O_4)_3,10H_2O$ *source.*

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