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FLUORESCENCE SPECTRA OF URANIUM, NEPTUNIUM, AND CURIUM

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Fluorescence of AmCl_3 and PuCl_3 in dilute solid solution in a matrix of crystalline anhydrous LaCl_3 has been reported previously.^{1,2} We have since observed the fluorescence of U^{+3} , Np^{+3} , and Cm^{+3} in a similar crystalline environment. Observed fluorescence lines and, for Cm^{+3} , absorption lines in the region 3000 to 8000 Å are listed in Table I. The fluorescence spectra were obtained by ultraviolet irradiation, as described previously,^{1,2} although for curium (~185 $\mu\text{g Cm}^{244}$ in 183 mg LaCl_3) the radioluminescence is sufficiently intense to be photographed on our 21-foot Wadsworth spectrograph.³ The radioluminescence spectrum, aside from its lower intensity, is identical with that obtained by ultraviolet irradiation. The strong fluorescence group at 4000 Å continued to fluoresce in absorption experiments, and therefore has not been observed as an absorption line by us. It is now known, however, that this appears as a peak in the absorption spectrum of aqueous Cm^{+3} .⁴

The fluorescence of NpCl_3 in LaCl_3 was observed at a concentration of ~0.1 atom % Np^{237} similar to the CmCl_3 preparation.

Because of the ready availability of natural uranium, solutions of UCl_3 in LaCl_3 up to 20 weight % were prepared.

Attempts were made to incorporate UCl_3 in NaCl , SrCl_2 , BiCl_3 , and MgCl_2 . Although some uranium appeared to dissolve in some of the crystals, none of the products was fluorescent.

Some interpretation of the observed spectra is possible. The ground state of Cm^{+3} is $^8\text{S}_{7/2}$, with insignificant splitting by the hexagonal crystalline field of LaCl_3 , so far as optical spectra are concerned. Multiplet structure therefore arises from crystal-field splittings of excited states. The three-component group of lines at 4600 Å therefore arises from a $J = 5/2$ level, the four-component group at 4000 Å from a $J = 7/2$ level, and the two doublets at 3830 Å and 3780 Å from $J = 3/2$ levels. The group at 4600 Å

probably arises from the ${}^6P_{5/2}$ level, that at 4000 Å from the ${}^6P_{7/2}$ level (although it may be the ${}^6I_{7/2}$), and the two groups with $J = 3/2$ from 6P , 6D , 6F , or 6G , or possibly from quartet S,P,D,F, or doublet P or D, are entering.

The ground state of Np^{+3} is 5I_4 . A preliminary analysis suggests levels at 60 and 110 cm^{-1} above the ground state. The level of 19870 cm^{-1} is not split and therefore has $J = 0$. The most reasonable assignment for this level is 5D_0 , although 3P_0 and 1S_0 are possible.

The doublet at 16070 cm^{-1} is split by 25 cm^{-1} . For this $J = 1$ level the possibilities are 5F , 5D , 5P , 3D , 3P , 3S , 1P .

The interpretation of the uranium fluorescence spectrum is more difficult than for Cm^{+3} or Np^{+3} . The splitting of the ${}^4I_{9/2}$ ground state appears to contain an interval of 180 cm^{-1} followed by a 20 cm^{-1} interval. Absorption-spectra observations suggest that there may be an additional level at about 25 cm^{-1} above ground.

Table I. Spectra of U, Cm, and Np in $LaCl_3$
(wavelength in angstroms)

<u>$CmCl_3$ in $LaCl_3$: Fluorescence</u>					
4606	4603	4588	4001	3995	3990
3984					
<u>$CmCl_3$ in $LaCl_3$: Absorption</u>					
3835	3830	3787	3776		
<u>$NpCl_3$ in $LaCl_3$: Fluorescence</u>					
6261	6252	6243	6232	6219	6208
5118	5060	5046	5030	4998	
<u>UCl_3 in $LaCl_3$: Fluorescence</u>					
6928	6915	6898	6812 ^b	6800 ^b	
6482 ^{a,b}	5513	5506	5452 ^b		

^aFluorescence line appears only at room temperature

^bAlso appears in absorption

References

1. Gruen, Conway, and McLaughlin, J. Chem. Phys. 24, 1115 (1956).
2. Cunningham, Gruen, Conway, and McLaughlin, J. Chem. Phys. 24, 1275 (1956).
3. M. Fred and D.M. Gruen (Argonne National Laboratory) (private communication) have observed this radioluminescence on a fast spectrograph, and confirm our results.
4. T.K. Keenan, P.R. Fields, W.T. Carnall, and D.C. Stewart (of Los Alamos Scientific Laboratory and Argonne National Laboratory) (private communication).