

## Study on the Separation Factor of Am-Cm in the Di-(2-Ethyl Hexyl) ortho Phosphoric Acid Extraction System

by  
Chin Taik Rhee  
Inha Institute of Technology, Korea

### HDEHP-HCl 系에 있어서의 Am-Cm 의 分離係數

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仁荷工科大学

李 鎮 濤

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#### ABSTRACT

The Separation factor of Am-Cm in HDEHP-toluene-HCl system was studied using discontinuous counter current extraction technique. The value was found as 1.31 which is much smaller than that of neighbouring elements in 4f series.

#### INTRODUCTION

Trivalent Am and Cm have many similarities as neighbouring elements in the sense of atomic number in 5f series as well as the mutual similarity between neighbouring elements in 4f series. There are many works reported<sup>(1-6)</sup> for the mutual separation of Am and Cm by ion exchange resin but no concentrated study on that subject by liquid extraction technique is known. It is well known that Di-(2-ethyl hexyl) ortho phosphoric acid (HDEHP) is one of the most effective (greater separation factor) solvent<sup>(7)</sup> for the mutual separation of 4f series elements. Some study for the mutual separation of 4f series elements by discontinuous counter current extraction technique with HDEHP was tried and available results were reported<sup>(8)</sup> for the mutual separation of Nd-Pm in which study more than 80% of pure Pm was recovered through 40 cells-60 organic batches operation. The author desired to compare the two series in the same extraction system.

#### EXPERIMENTAL

**Source of materials:** HDEHP was purified according to the method given by D. F. Peppard *et al.*<sup>(9)</sup>

The purity was checked by measuring the distribution ratio (Kd)\* and the acid dependency of Eu. All the chemicals including toluene used as diluent were analytical or chemical pure grade. Eu<sup>152</sup>, <sup>154</sup>Pu<sup>239</sup>, Am<sup>241</sup> were received from Radio Chemical Center, England.

**Irradiation:** An aliquot amount of nitrate solution of Am<sup>241</sup> was taken on a small piece of polyethylene sheet and dried under infrared lamp. This target was irradiated as usual solid sample in the rotary specimen rack of TREGA reactor ( $\phi \sim 10^{12}$ ) in the Atomic Energy Research Institute, Korea for more than three months. After irradiation, the Am-Cm mixture was recovered in nitric acid solution by dissolving out.

**Radioassay:** Gamma counting was carried out with NaI(Tl) well type scintillator. For  $\alpha$  or gamma spectrum, C-4-25-0.2 solid detector (supplied by RCA Victor, Canada) or 3" x 3" NaI (Tl) scintillation detector connected with RCL 256 channel analyser was used. The peak analysis of  $\alpha$  energy spectrum was carried out with the aid of the spectrum of Pu<sup>239</sup> and Am<sup>241</sup>. Since the counting rate of  $\alpha$  activity is very sensitive to the plating condition, the activity in organic sample was mounted on plate after back extraction in an aqueous solution of adequate acidity.

**Extraction apparatus:** The extractor used was a

conventional Craig type (supplied by Tajiri, Japan). The capacity of each cell for heavier phase was 10 ml. Except the shaking stage which is operated and controlled automatically by speed controller and timer, all the stages were operated by manual handling. 40 cells (out of its 60 maximum loading capacity) were used for the present work.

**Extraction procedure:** All the pre-treatment of the cell prior to real operation was done as the previous work<sup>(8)(9)</sup>. After filling up all equilibrating cells with 0.041 N HCl solution, an aliquot amount of irradiated sample which was dried under infrared lamp was transferred to the first cell by dissolving out with the acid solution in that cell. Five ml portion of 40 organic batches (10% (v/v) HDEHP-tol) were supplied to the first cell one by one successively. Extraction and transfer cycles were repeated after completion of the above organic solvent supply until the last organic fraction was withdrawn from the last cell. Each shaking period was six minutes.

## RESULTS AND DISCUSSION

Fig. 1 shows the measured  $\alpha$  spectrum of Am-Cm mixture. The energy of 6.11 Mev calibrated by 5.15 Mev peak (Pu<sup>239</sup>) and 5.47 Mev peak (Am<sup>241</sup>) is fairly good fit to the reported energy value of Cm<sup>242</sup>. Such separated peaks were used for the calculation of each activity fraction in the sample of mixed activity. The results of counter-current operation are summarized in Fig. The aqueous fractions are numbered as 1 A and 40 A for the first and last cells respectively. The organic fractions are numbered as 1 s, 2s, ... successively from the first effluent. The total  $\gamma$  activity of each fraction was analyzed by dividing in proportion to each  $\alpha$  activity in each fraction. The maximum peaks of Am and Cm are observed in 22A and 29A respectively. Thus the distribution ratio is calculated by the aid of the equation derived by E. Hecker as follows.

$$Kd = \frac{\gamma_{max} + 0.5}{m} \times 2$$

Where,  $\gamma_{max}$ : number of aqueous fraction of maximum activity

Observed,  $m$ : number of total cell

Thus,  $Kd(\text{Am}) = 1.125$ ,  $Kd(\text{Cm}) = 1.475$

The separation factor =  $\frac{1.475}{1.125} = 1.31$

This value of separation factor is fairly close to the

value of 1.3 obtained by R. D. Raybarz<sup>(11)</sup> for 2-ethyl hexyl phenyl phosphoric acid-diethyl benzene-HCl system and much smaller than that of lanthanon series ( $\sim 2.5$ ) for the same HDEHP extraction system.

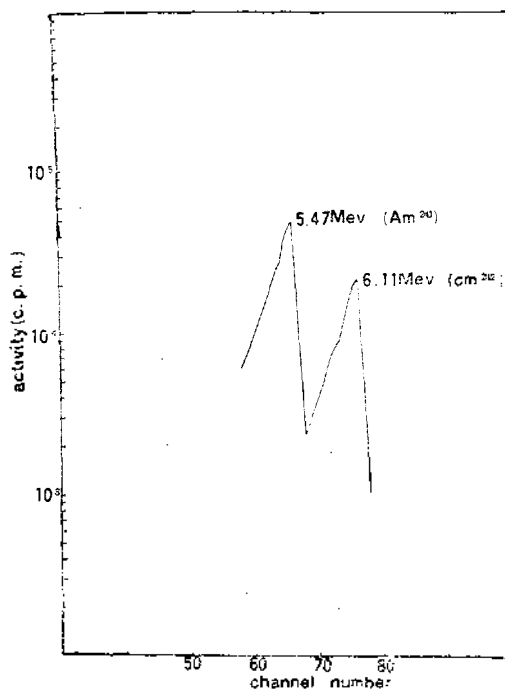


Fig. 1.  $\alpha$  spectrum of Am<sup>241</sup> and Cm<sup>242</sup>.

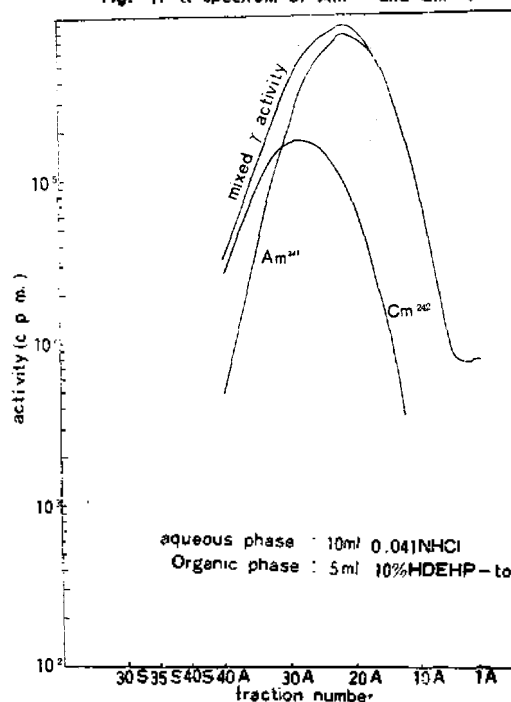
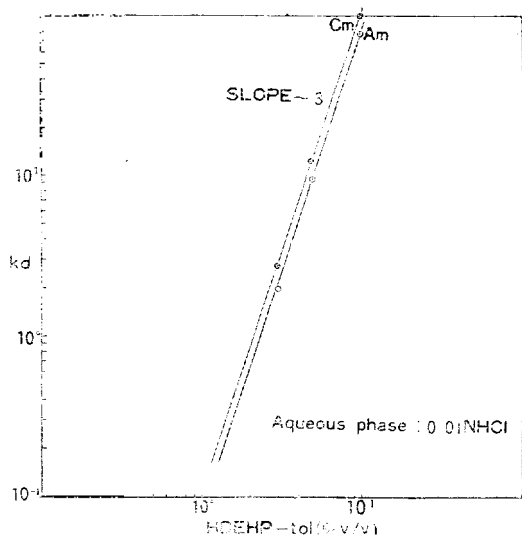
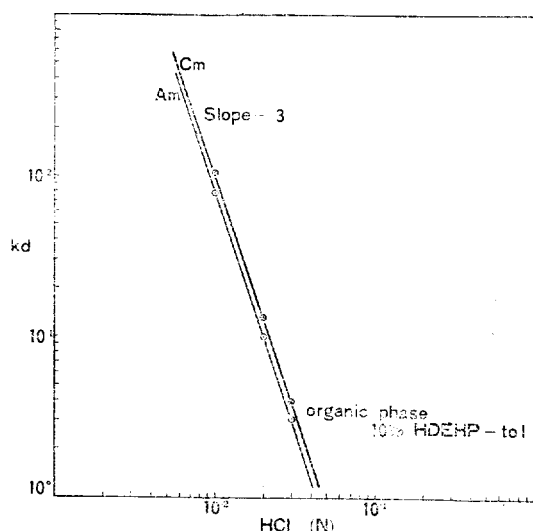


Fig. 2. Distribution curve of Am<sup>241</sup> and Cm<sup>242</sup> activities.

Fig. 3. Activity dependency of  $K_d$  for Am and Cm.Fig. 4. Solvent dependency of  $K_d$  for Am and Cm.

As shown in Fig 3 and 4 which are drawn by plotting the  $K_d$  value obtained by batch extraction study, the distribution ratio of Am or Cm shows a direct third power dependence upon the concentration of HDEHP in the equilibrated organic phase and an inverse third power dependence upon the hydrogen ion concentration in the equilibrated aqueous phase as same as the case of lanthanons. The value of distribution

抄: 不連續向流分配裝置를 利用하여 HDEHP-tol-HCl系에 있어서의 Am과 Cm의 分離係數를 測定해 보았으며, 얻어진 1.31의 값은 4f系列에 있어서의 그것 보다 훨씬 작음을 알았다.

ratio of Am is found fairly close to that of Nd in 4f series.

These results show us that the elements in the two inner transition series, 4f and 5f, in same trivalent state form same extractable species but the formation or stability constant which is depends on the atomic number and shows fairly larger difference in 4f series has relatively smaller difference in 5f series.

One of the imaginable reason for the difference between the two series the difference in degree of availability of orbitals in hybridization which is larger in 5f series than 4f series

### SUMMARY

The distribution ratio of Am and Cm has a direct and inverse third power dependence upon the hydrogen ion and solvent concentration as the case of the trivalent lanthanous but the separation factor is found to be 1.31 which is much smaller than that in lanthanon series. The  $K_d$  value of Am is fairly close to the value of Nd in 4f Series.

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The distribution ratio was defined as the ratio of counted activity of organic phase to that of aqueous phase under equal volume base