H A

UCRL-ID-126660

# Critical Review of Progress Report on Reactive Matrix Isotope Separation by E. Catalano, E. Lee, and M. Schwab

E. Catalano

RECEIVED
APR 14 1997
OSTI



August 19, 1975

Lawrence Livermore National Laboratory



Work performed under the auspices of the U.S. Department of Energy by the Lawrence Livermore National Laboratory under Contract W-7405-ENG-48.

## DISCLAIMER

**Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.**

## DISCLAIMER

[redacted]

[redacted]

COCB AUG 21 1975

59/258

[redacted]

August 19, 1975

M E M O R A N D U M

L I S

TO: Harry Rizzo

FROM: E. Catalano

SUBJECT: Critical Review of Progress Report on Reactive Matrix Isotope Separation by E. Catalano, E. Lee, and M. Schwab

---

Professor John L. Margrave is a consultant for us. Professor Margrave is Dean of the Graduate School, Department of Chemistry, Rice University in Houston. He has had wide experience in the fields of high temperature chemistry, fluorine chemistry, and chemical reactions carried out by low temperature matrix isolation techniques. We were pleased to have Professor Margrave critically review our paper. Enclosed are his comments.

You will notice that he agrees with us as to the important criteria that still have to be met if our process is to be a viable one. But also notice that he suggests a strong effort along the lines of investigating other candidate chemical systems. His suggestions are well thought out and we will be taking them into consideration.

He has made a very valuable suggestion for a method of pre-cooling $UF_6$ for the matrix isolation which we hope to implement.

S. R.

E. Catalano

...Changed to:
UNCLASSIFIED
(...assification level or indicate Unclassified)
COCCA-75-9 6/7/96 (date)
(...ge in classification, e.g., the memorandum number.)
10/1/96 (date)
(...e of person making the change)
...ne Barrow 10/4/96 (date)
(...n verifying this is the correct document or model)

EC:de

Distribution:

| | |
|---|---|
| Copy 1 of 9A | H. F. Rizzo |
| Copy 2 of 9A | G. D. Dorough |
| Copy 3 of 9A | J. W. Dubrin |
| Copy 4 of 9A | E. L. Lee |
| Copy 5 of 9A | A. Maimoni |
| Copy 6 of 9A | L. W. Roberts |
| Copy 7 of 9A | M. Schwab |
| Copy 8 of 9A | E. Catalano |
| Copy 9 of 9A | File |

[redacted]

Classification (...
U...
(Insert appropriate c...
by authority of R2D2-...
(Authority for chan...
by
(Signature...
verified by
(Signature of perso...

cm 8/21/75

LL-793

Comments on Draft #5, "Reactive Matrix" Work Paper

J. L. Margrave

1. This paper presents an intriguing introduction to some real possibilities for uranium isotope separation using matrix techniques.

2. There are three critical problems which should be resolved to validate the approach, as soon as possible:

   (a) The photo-induced reactions:

$$(NO + UF_6)_{matrix} \xrightarrow{h\nu} (NO^+, UF_6^-)_{matrix}$$

$$(CO + UF_6)_{matrix} \xrightarrow{h\nu} (CO\,F_2 + UF_4)_{matrix}$$

   must be shown to occur -- initially with a broad band of IR ($600 \pm 100$ cm$^{-1}$) which will show no isotopic discrimination.

   (b) A mixed matrix of $UF_6$ + NO, unreacted, must be prepared; also, $UF_6$ + CO matrices, or $UF_6 + X_e$ matrices.

   (c) A monochromatic source of ∿620 cm$^{-1}$ radiation must be found.

3. In order to develop this approach fully, one needs:

   (a) A better chemical and structural and thermal understanding of $NOUF_6$, $NOUF_7$, etc.

      (1) Are these charge-transfer, ionic solids?

      (2) What are crystal energies, heats of formation?

      (3) How do these compounds melt, decompose and vaporize?

   (b) Studies of $UF_6$ in $F_2$, Ne, Ar, Xe, CO, $N_2$, and NO matrices to establish optimum matrix gases, to get best line shapes, and to choose best reaction.

   (c) Studies of other possible reactions of $UF_6$ (in addition to NO and CO), for example:

      (1) $Xe + UF_6 \longrightarrow (Xe^+, UF_6^-)$

      (2) $Li + UF_6 \longrightarrow (Li^+, UF_6^-)$

      (3) $\begin{Bmatrix} CF_2 \\ SiF_2 \end{Bmatrix} + UF_6 \longrightarrow UF_4 + \begin{Bmatrix} CF_4 \\ SiF_4 \end{Bmatrix}$

      (4) $O_2 + UF_6 \longrightarrow (O_2^+, UF_6^-)$

4. In terms of the process, for $M/UF_6 \approx 1000$,

$NO_xF$
where $M = NO$ or $Xe$ or $O_2$ or $Li$

$$(M + UF_6)_{matrix} \xrightarrow[20K]{h\nu} \left[M + {}^{235}UF_6 + {}^{238}UF_6\right]_{matrix}$$

$$\downarrow \text{warm-up}$$

$$(M\,{}^{235}UF_6)_{solid} + {}^{238}UF_6(gas) + M \xleftarrow[\text{temp}]{\text{room}} \left\{(M^+, {}^{235}UF_6^-) + {}^{238}UF_6\right\}_{matrix} + \text{excess } M$$

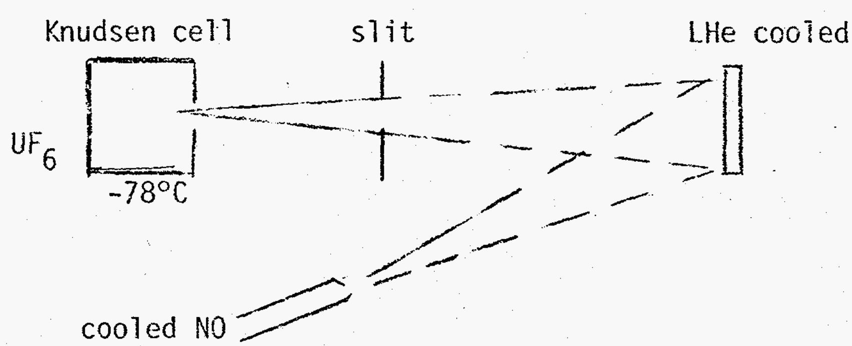
$$\downarrow$$

$(NO^+, UF_6^-)$
$(NOF^+, UF_7^-)$
$(Li^+, UF_6^-)$
} all known, stable solids; form spontaneously at room temperature

$(O_2^+, UF_6^-)$
$(Xe^+, UF_6^-)$
predicted to be stable solids, like $PtF_6$ analogs. If not currently known, these should be prepared and characterized.

## CONCLUSIONS AND SUGGESTIONS

1. Need detailed studies of $UF_6$ in Ne, Ar, Xe, $N_2$ matrices.
2. Need system for making $NO + UF_6$ matrix:




could "cryopump" cold $UF_6$ from $CO_2$-cooled Knudsen cell to LHe-cooled surface and CO-deposit cooled NO to get unreacted $NO/UF_6$ matrix

[redacted]

 [redacted]

3. Need to explore Xe + $UF_6$ system.
   (a) is reaction observed at room temperature?
   (b) is reaction observed at low temperature?
   (c) is $XeUF_6$ stable?
4. Could use laser raman to characterize reaction products.

[redacted]