

Destruction of High Concentration Nitric Acid by a Continuous Denitration by Formic Acid - Electrolytic Trimming of Residual acid

150, 305-600

가

2.0 M

0.1M

Abstract

This work has improved the conventional batch denitration by formic acid which has been used for controlling the acidity of solution for the solvent extractions to partition the long lived-radionuclides from the high level radioactive liquid waste. The characteristics of destructions of nitric acid and formic acid and their destructive products in a continuous denitration process combining the continuous denitration by formic acid and the residual acid-trimming suggested in this work was evaluated. The new continuous denitration process combining the denitration by formic acid and residual acid-electrolytic treatment could control treat continuously a feeding nitric acid of 2.0 M to below about 0.1 M.

1.

(HLW:high-level radioactive Waste)

(partitioning-transmutation)

가

[1,2]

HLW

(long-lived nuclides : Am, Cm, Np, Tc),

(Cs, Sr),

(Pd, Ru,

Rh) (fission product) ,
(short-lived nuclides)

가

[1-6],

JAERI

[3,4]

Zr, Mo

[3,4,6-8]

가

가

가

가

가

가

1.5

2.5

0.5 M

가

[4,6,9]

가 1.5

1.5

가

가

가

, 2

0.05 - 0.5M

가

1.5

- 가
.[9]

가

2.

가 Fig.1
20 mm 50, 80, 100, 125 ml
Pyrex column 84°C - 96°C
thermocouple column heating tape
가
NOx damper tank
NOx 가 flow meter가
2.0 M 2.0 M
0.5 - 2 ml/min syringe pump , 25.9
M 1.5 - 2.0 / syringe pump
18ml
3
가
(OH^{*})
DSA (dimensionally stable anode) 가
,
(multi-coating layer) 가 mesh IrO₂ [10-13],
mesh Ti 23 cm², 5 mm
, 18 ml
Auto-titrator (Kyoto Electronics AT-400)
TOC (Shimadzu TOC-5000A),
, Ion chromatograph (Dionex ICS-90)

3.

가
 Fig.2 가 125 ml
 / 2.0
 가
 / 가 2.0 M
 150 ml가 250 ml 가 2.0
 25.9 M 17 / 가 2.0
 0.5 M 1.5
 M 30
 가

가 가
 5 M
 (HNO₂)
 hyponitrous radical(HNO*) format radical (HCOO*)

[5,6,9]

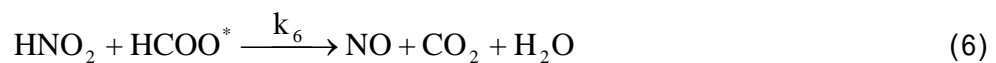
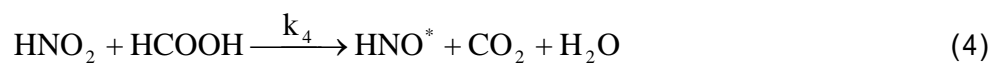
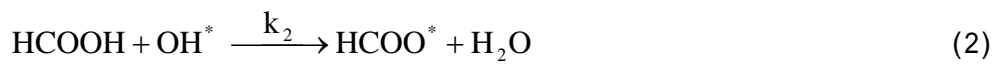
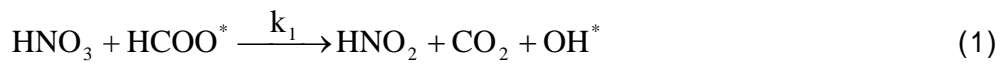


Fig.3 / 가 2.0 가
 가 Fig.2 Fig.3

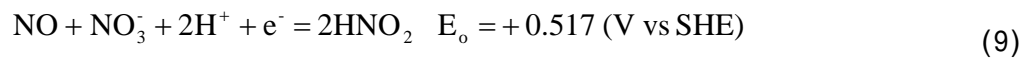
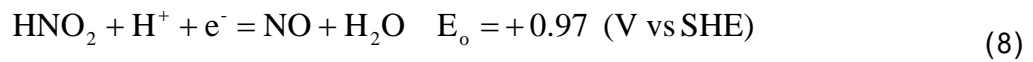
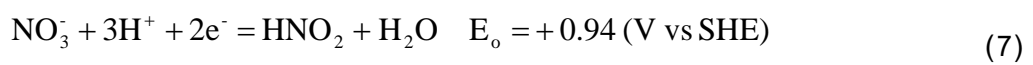
Fig.4 / 125 ml 가 / 가 1.5
 가 .[9] 가

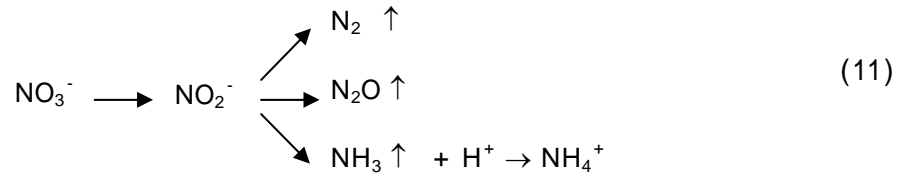
Fig.5 / 가 2.0 125 ml 가 가
 가 . 98°C

Fig.2 - Fig.5 가 가
 가 가

Fig.6 (A) IrO₂-Ti 1.0 M HCOOH + 0.5 M HNO₃ 60 mA/cm² 가
 . 60 0.3 M, 0.5 M 가 ,

(7) (8) H⁺ NO HNO₂ . [14,15]
 (11) , NO_x . [16,17]
 pH .





radical (OH^*) (12) . [10-13] OH^* 가 hydroxyl



. [18]

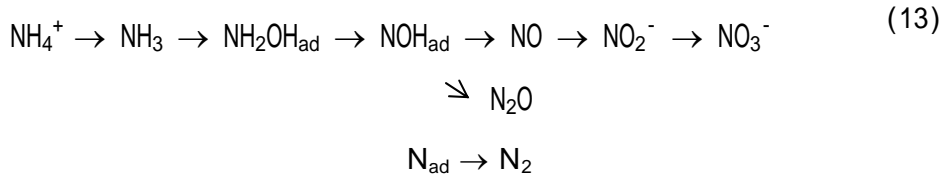


Fig.6 (A)

가

(Nafion 424) 가 IrO₂-Ti

1.0 M HCOOH + 0.5 M HNO₃

가 Fig.6 (B) (C)

(11)

가 가 (5)

가

Fig.6(A)

. Fig.6(B)

가
가

(11)

Fig.6(C)

가

가

가 . Fig.7

IrO₂-Ti 가

9.33x10⁻³ M/(hr.mA/cm²)

1.37x10⁻² M/(hr.mA/cm²) 가 50 %

가 80 mA/cm² 1 1.0 M 가

, 0.6 M 1.6 M 1

Fig.4 / 1.5

/ 1.5

가

Fig.8 가 가 가 가 가

50°C

2.0 M / 1.75

96°C 50°C

Fig.9 3 , ,

가 2.0 M

1.5 M , 3 0.69 M, 0.2

M, 0.05 M 가 1.0 M 가

0.5 M 가

10⁻³ M , Fig.6

NOx 가

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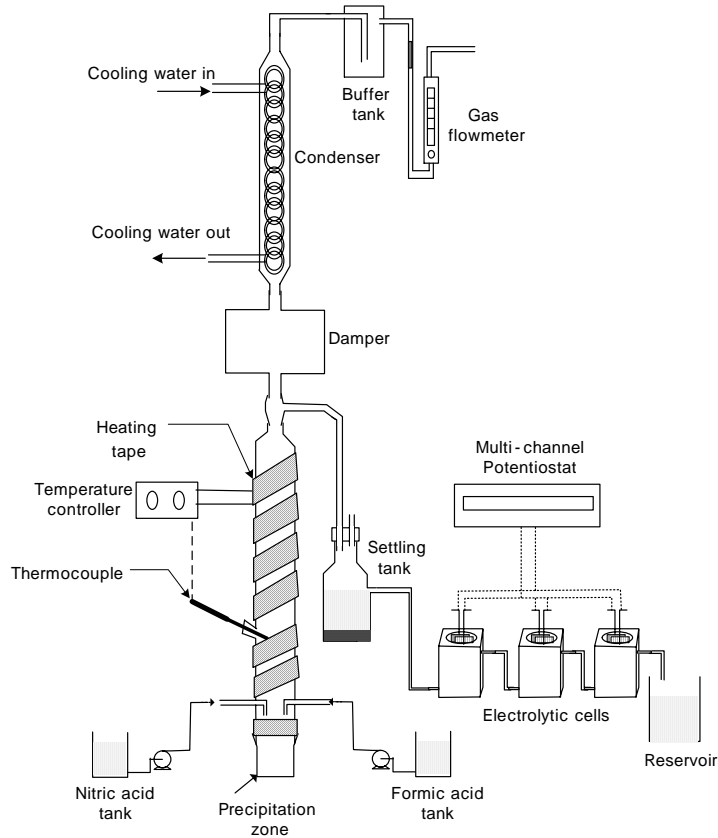


Fig.1. Experimental apparatus consisted of chemical and electrolytic systems for denitration.

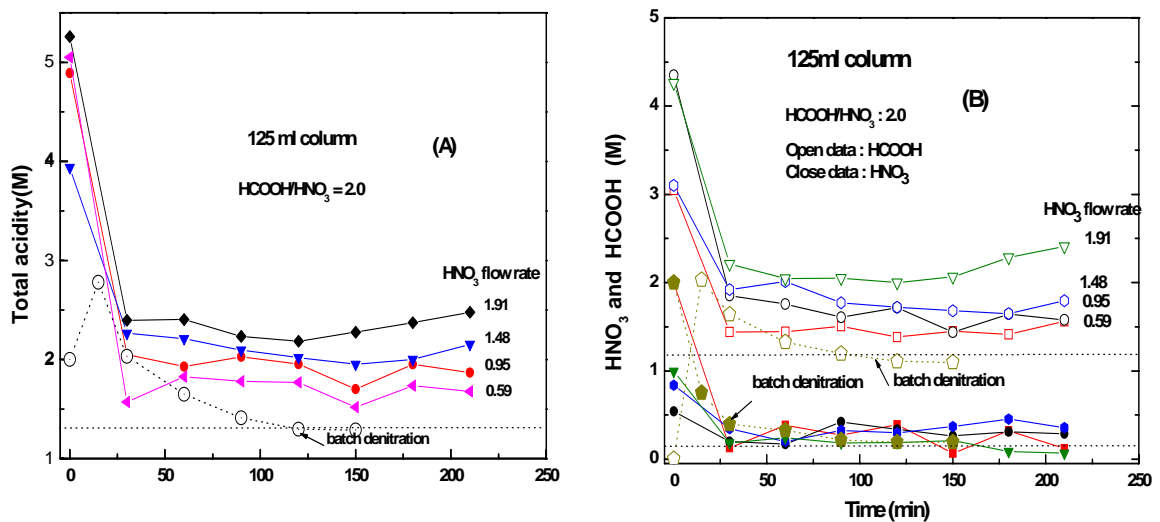


Fig.2 Changes of total acidity (A), nitric acid and formic acid (B) in 125 ml column at several nitric acid flow rates at a mole ratio of formic acid to nitric acid of 2 with time.

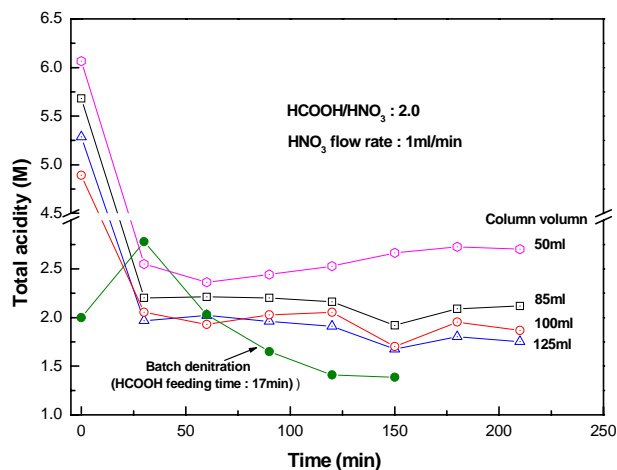


Fig.3 Change of total acidity in several columns with time

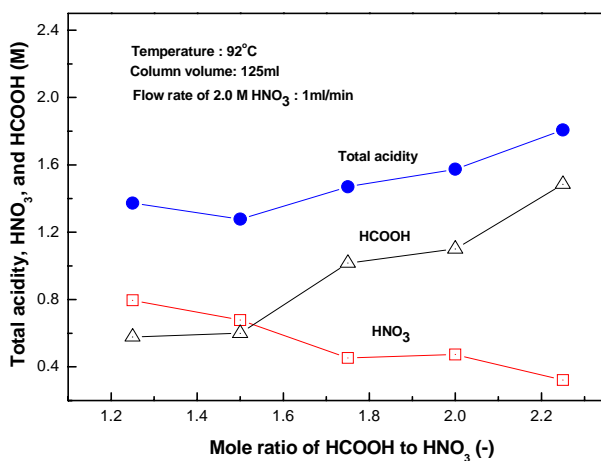


Fig.4 Changes of total acidity, nitric acid and formic acid in 125 ml column at nitric acid flow rate of 1 ml/min with mole ratio of formic acid to nitric acid.

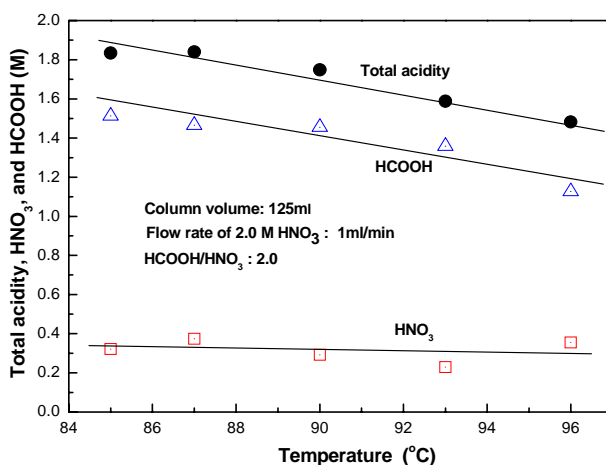


Fig.5 Changes of total acidity, nitric acid and formic acid in 125 ml column at a mole ratio of formic acid to nitric acid of 2 with denitration temperature.

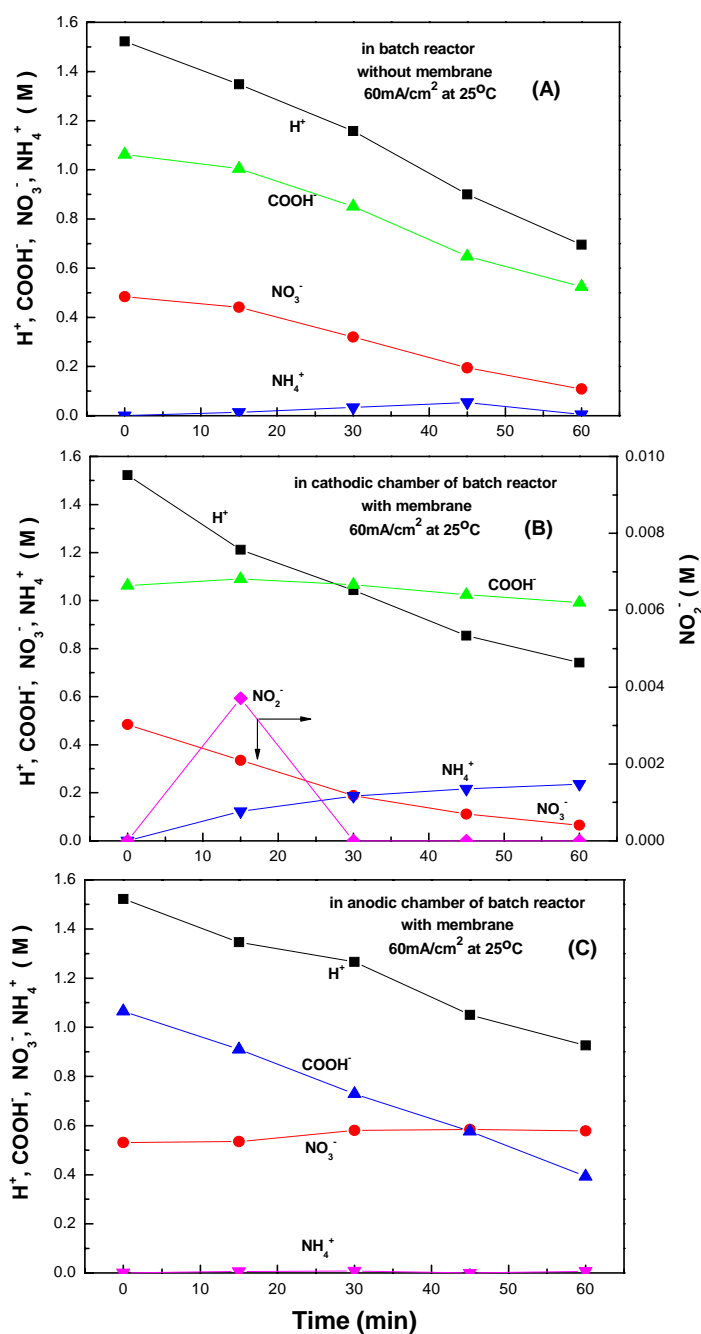


Fig.6 Changes of proton, nitrate, nitrite, formate, and ammonium ions with time in batch electrolytic reactor without membrane, and in cathodic and anodic chambers of batch electrolytic reactor with membrane at 60 mA/cm² with a mixture of 0.5 M nitric acid and 1.0 M formic acid.

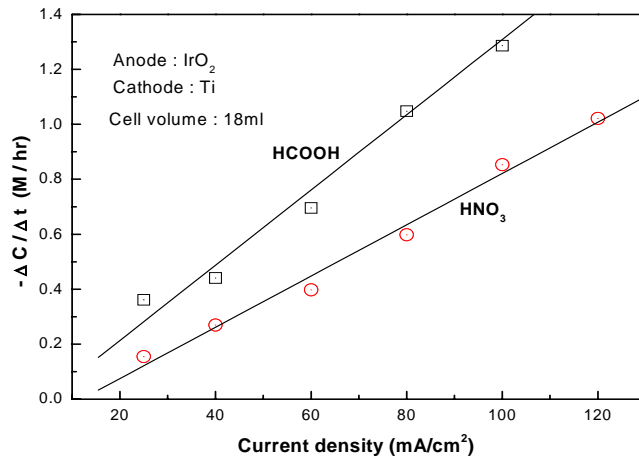


Fig.7. Electrolytic destruction rates of nitric acid and formic acid by an IrO₂-Ti batch cell with current density.

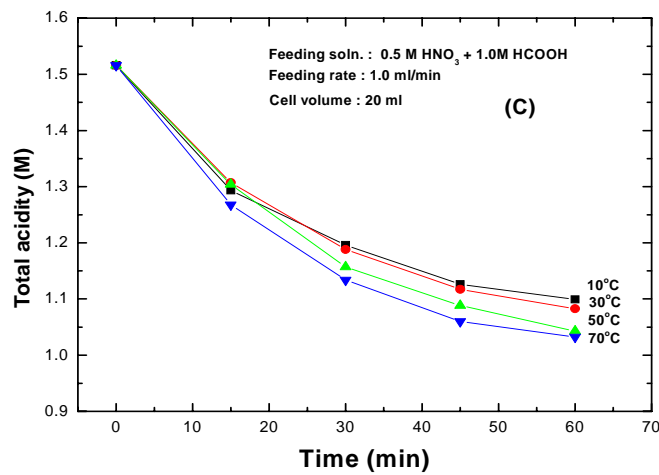


Fig.8 Changes of total acidity in a solution of 0.5 M nitric acid and 1.0 M formic acid in an IrO₂-Ti batch cell at several electrolytic reaction temperature with electrolytic time.

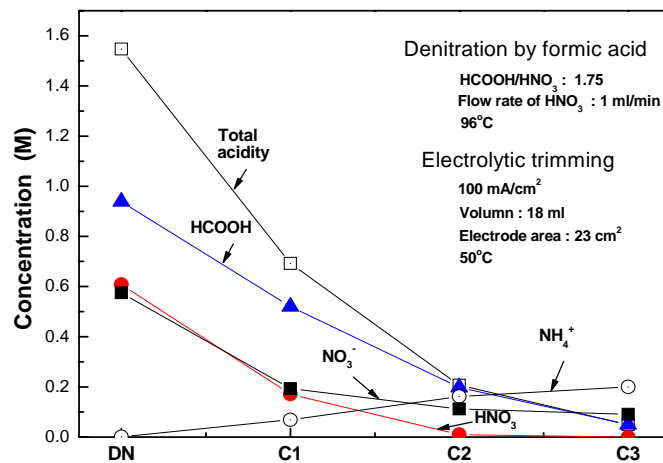


Fig.9 Changes of several species at each step of the continuous denitration by formic acid and the electrolytic cells.