

## 유동층의 공업적 이용

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## Industrial Applications of Fluidized Bed

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### 요 약

최근에는 流動層에 대한 應用研究가 活潑하다. 本報(第二報)에서는 流動層의 應用面에 대한 レビュー 몇개의 流動層反應器(特殊型)에 대한 特性을 살펴보고자 한다.

### Abstract

In recent years some noteworthy developments and the practical applications of fluidized beds to industry have been reported as the significance of fluidized bed has been recognized more and more. In this part II, recent advances on applied parts of fluidized beds are reviewed and characteristics of some special types will be discussed.

### 1. 서 론

1970년 이래 流動層에 대한 模型化가 盛行하다가 최근에는 그 應用面에 대한 研究, 특히 石炭, 重質油, 廢棄物의 燃燒에 의한 에너지化 및 氣體化, 鐵鑛石의 直接還元, 放射性廢棄物의 處理, 廢熱回收등 에너지回收과 環境分野에 대한 研究가 두드러지게 進行되어 지고있다.

流動層<sup>2)</sup>, 噴流層<sup>4)</sup> 및 傳熱<sup>1)</sup>에 관한 單行本이

發刊되었고, 流動層에 대한 國際會議가 Toulouse (1973), California (1975)에서, 反應工學에 대한 國際심포지움이 歐美에서, AIChE의 年會등에서 각각 發刊된 Proceedings<sup>3,7)</sup> 또는 Symposium Series<sup>11~17)</sup>가 있다. 流動層燃燒에 관한 London<sup>8)</sup> 및 Verginia<sup>9)</sup> 國際會議(모두 1975년)의 Proceedings, 廢棄物의 에너지로의 變換方法에 대한 Montreaux 國際會議의 Proceedings<sup>19)</sup>가 發刊되었다. 그리고 AIChE年會에서도 大氣汚染과 廢

藥物處理의 部會가 있었고, 流動層技術이 많이 포함된 Symposium Series<sup>11~8)</sup>가 發刊되기도 했다.

本報에서는 第 I 報에서 說明한 바 있는 模型化나 氣泡에 관한 論文 以外の 重要文獻을 대략 分類해서 뒷면에 記載하고 流動層操作上으로 보아 特殊한 것 몇개의 特性을 살펴보고자 한다.

## 2. 流動化機構 및 熱—物質傳達

流動化開始速度는 流動層反應器 操作上 기본적인 것이지만 그 測定이 그리 容易하지는 않다. 특히 石炭燃焼 또는 氣體化 反應裝置, 기타 熱分解裝置로서 流動層을 應用하러니 高溫 高壓下에서 操作하게 되며 粒子直徑과 比重差가 있는 混合系를 處理하게 되므로 이 流動化開始速度는 큰 影響을 주는 因子가 되고 있는 것이다. Rowe<sup>25, 28)</sup>는 多成分系粒子混合物의 流動化開始速度를, 其中 큰 粒子의 重量分率을 基準으로理論式을 제안하였고 Barnea-Mednick<sup>21)</sup>는 Barnea-Mizrahi<sup>22)</sup> 相關式에 混合系中 各粒子別 體積分率을 고려한 關係式을 제안한 바 있다. 高壓下의 流動化開始速度에 관하여는 Broughton<sup>24)</sup> 및 Mii<sup>39)</sup> 등의 研究가 있다. Broughton은 流動化開始速度에 影響을 주는 原因은 分散板 上下의 溫度差에 의한 分級이라고 說明하고 있으나 더 正確한 實驗値가 必要한 실정이다. Brodhust-Becker<sup>23)</sup>는 여러가지 因子를 바꾸어가며 多角적으로 시험하여 流動化開始速度보다는 氣泡發生最小速度를 基準으로 함이 더 有利하다고 발표한 바 있다. 白雲石 中에서 石炭을 燃焼할때, 어떤 廢棄物을 流動化되고 있는 모래中에 投入시켜 燒却시킬 때나, 粒子直徑과 密度가 큰 差가 있는 物質을 한꺼번에 流動化시킬 경우 層內에선 分級이 일어나게 된다. 이때 適當한 混合程度에 따라 裝置燃焼率을 크게 좌우하게 된다. 또 石炭으로 부터 생기는 灰分을 溢流除去 시키느냐 混合殘留케 하느냐 하는 것은 操作條件上의 문제일 것이다. 이 分級에 대하여는 Rowe<sup>99)</sup>가 그 기초적 研究를 시행하였고, jetsam (또는 flotsam)이란 새 用語를 사용하여 模型<sup>94)</sup>을 제안한 바 있다. Chen-Keairns<sup>93)</sup>는 石炭氣體化및 燃

燒行程에 대해 광범위하게 시험한바도 있다. 이것은 마치 氣—液間의 相平衡과 유사하여 流動化狀態의 粒子層과 그것을 固定層으로 있게 할 때의 粒子層 相互間의 平衡關係를 나타내는 흥미로운 結果를 얻었다. 多成分混合物에 대한 分級研究<sup>96, 102)</sup>도 다소 있다.

傳熱에 관한 것은 單行本<sup>1)</sup>뿐 아니라 Gutfinger-Abuaf<sup>107)</sup>, Mori<sup>114)</sup>, Sparrow et al<sup>117)</sup>, 및 Yoshida<sup>20)</sup> 등의 報告가 있고 McGaw<sup>130)</sup>는 횡단류 流動層 熱交換器內 傳熱을 다루었다. 그리고 Zabrotsky<sup>120)</sup>는 層內表面間 傳熱을 다룬 바 있다. 物質傳達에 관하여는 Yokota<sup>136)</sup>, Yoon-Thodos<sup>138)</sup> 등이 발표한 바 있고, Dwivedi-Upadhyay<sup>139)</sup>는 지금까지 발표된 자료를 재분석하고 상관關係式을 구하여 폭넓게 이루었다.

飛末流出<sup>139~144)</sup>, Slugging<sup>156~161)</sup>, 噴流層<sup>162~179)</sup>에 대한 것은 뒷면에 分類된 참고문헌을 보기바란다. 分散板에 있어서는 氣泡徑과 관련되므로 設計上<sup>150, 151, 366)</sup> 대단히 중요하다.

## 3. 流動層의 物理的 應用

중요한 것으로는 乾燥에의 應用<sup>205, 206, 208, 211, 225)</sup>, 被覆, 凝集, 造粒에의 應用<sup>212, 215, 218~224, 227, 229)</sup>, 및 熱交換에의 應用<sup>209, 211, 217, 220, 226, 231)</sup> 등으로 볼 수 있다. Krause, Slanky의 研究는 放射性廢棄物에의 應用이고, Keairns는 高速增殖爐의 冷却에 쓰인 高溫 Na의 熱回收方法에 대한 研究로서 傳熱係數가 2,000kcal/m<sup>2</sup>.hr.°C 以上인 것으로 報告되었다. 그리고 Donsi-Russo는 流動層을 急速冷却器로 쓸 수 있음을 시사하였다.

## 4. 石炭—油類燃焼 및 固體廢棄物 處理에의 應用

서론에서 논한 바와 같이 이 계통의 論文이 압도적으로 많다. 그 內容에 따라 보면 燃焼<sup>232~238)</sup>, 脫黃이나 再生<sup>239~242)</sup> 분야의 研究가 대부분이다. 石炭의 氣體化관계는 Kunii<sup>252)</sup>, Osa<sup>256)</sup> 및 Perry<sup>257)</sup> 등의 研究가 있고 重質油의 氣體化에 있어서는 Klass<sup>264)</sup>의 解說이 있다. 氣體燃焼를

취급한 것은 Davis<sup>233)</sup>와 Singh<sup>237)</sup> 뿐이다. 流動層보일러의 材質에 관한 論文은 Cooke<sup>243)</sup>, Goldberger<sup>244)</sup> 및 McNab<sup>245)</sup> 등이 있다. 그리고 Thring<sup>238)</sup>은 Stirling 熱機關에 熱源의 공급 방법으로서 傳熱係數가 높고 ( $400 \sim 750 \text{ W/m}^2 \cdot ^\circ\text{K}$ ), 等溫 ( $850 \sim 1100^\circ\text{C}$ ) 操作이 가능한 流動層燃焼에 대한 研究에서 模型化와 應用可能性을 보였다.

固體廢棄物處理에 관한 것은 集塵, 타이어<sup>303)</sup>, 프라스틱<sup>304, 309, 312)</sup>, 스러지<sup>305, 313)</sup> 등의 工業廢棄物處理時 熱의 回收利用에 관한 것이 대부분이다. 그리고 Robert-Yates<sup>239)</sup>는 연도가스내  $\text{SO}_2$ 를 除去하는데 應用하기도 하였다.

## 5. 冶金에의 應用 및 流動層電極

流動層反應器는 鑛石分離에 대단히 有利함으로 鑛石의 乾燥, 煨燒, 焙燒, 抽出 및 精製등의 분야에 광범위하게 應用되어지고 있다. Doheim<sup>281)</sup>가 이 분야의 全般的인 利用에 대한 解説을 하였다.

鐵鑛石還元에의 應用<sup>279, 290, 291, 292)</sup> 및 우라늄 製鍊에의 應用<sup>272, 273, 286)</sup>이 중요한 것들이다. HIB나 FIOB 工程<sup>300)</sup>은 각각 年間 100萬ton, 40萬ton 규모로 제철공장이 운전되고 있다. 그리고 石灰石의 分解, 모리브테나이트 焙燒<sup>285, 280)</sup>, 일테나이트의 鹽素化<sup>299)</sup>, 電導性流動粒子間 電極插入으로 直接加熱에 의한 金屬製鍊<sup>282, 294)</sup>, 活性炭의 賦活<sup>297)</sup>, 觸媒再生<sup>302)</sup> 등의 研究를 볼 수 있다.

그리고 流動層電極<sup>343~359)</sup>에 관한 것은 前報에서 설명되었으나 그 文獻의 소개가 未洽하여 더 추가한다.

## 6. 특수流動層의 特性

### a. 眞空流動層

氣體流動層이 거의 모두 常壓下에서 操作되지만 減壓下에서 操作하게 되면 熱에 민감해 저서 流動化, 壁—粒子間의 傳熱 및 粒子—流體間의 物質移動에 관한 特性이 增大된다는 사실에 근거를 둔 減壓狀態를 眞空流動層이라 한다.

減壓下에서의 流體의 거동은 常壓下 操作에서나 거의 같으며 流動化效率은 形狀係數, 粒子徑이 적을 수록 나쁘고 공급되는 氣體의 流動化에 영향을 받지 않는다. 각粒子의 流動化效率은 表 1.에 나타났다.

표 1. Efficiency of fluidization<sup>305)</sup>

| Samples      | Avg. Particle dia. (mm) | Efficiency |
|--------------|-------------------------|------------|
| Glass bead   | 0.108                   | 0.27       |
| "            | 0.226                   | 0.25       |
| "            | 0.295                   | 0.20       |
| Glass powder | 0.115                   | 0.52       |
| "            | 0.214                   | 0.44       |
| "            | 0.289                   | 0.36       |
| Sand         | 0.239                   | 0.26       |
| "            | 0.327                   | 0.23       |
| Coke powder  | 0.259                   | 0.28       |
| Al powder    | 0.196                   | 0.39       |
| Acryl powder | 0.083                   | 0.56       |

眞空流動層에서 壁—粒子間의 傳熱特性에 영향을 미치는 因子에 대해 살펴보면 다음과 같다

#### i) 最少安定化流量

그림 1에서 보는 바와 같이 壁—層間의 傳熱係數는  $\frac{G}{G_{mf}} \geq 10$  범위에서는 거의 일정한 값을 가지나  $\frac{G}{G_{mf}} < 10$  범위에서는 流量이 적어짐에 따라 傳熱係數,  $h_w$ 도 감소한다.

#### ii) 壓力의 영향

$h_w$ 는 壓力이 낮아짐에 따라 감소한다. 그 이유는 壓力이 낮은 범위에서 流體가 粘性이 變해서 유효한  $\frac{G}{G_{mf}}$ 가 감소하기 때문에 流動化狀態가 劣化하여  $h_w$ 가 현저한 감소를 나타낸다. 壓力이 낮아지면  $\frac{G}{G_{mf}} \leq 1$ 로 떨어지게 되며 따라서  $h_w \leq 10$  정도로 낮아진다.

#### iii) 粒子徑의 영향

常壓에서  $h_w$ 에 대한 粒子徑의 영향은 Vreedenburg<sup>2)</sup>의 실험결과에 따르면 다음과 같다.

$$h_w \propto d_p^{-0.56}$$

#### iv) 氣相의 영향

$h_w$ 에 기여하는 因子로는 粒子—粒子間, 粒子

壁間, 粒子—氣體間, 壁—氣體間의 傳熱, 輻射 등을 생각할 수 있다. 이와 같은 특성을 살려 粒子의 乾燥나 流動化粒子를 傳熱媒體로한 沈澱物의 乾燥에 利用할 수 있으며, 특히 신속한 氣相分解反應, 固體熱分解反應등에 應用可能하지만 減壓裝置로 인한 操作上的 문제점이 있게 된다.

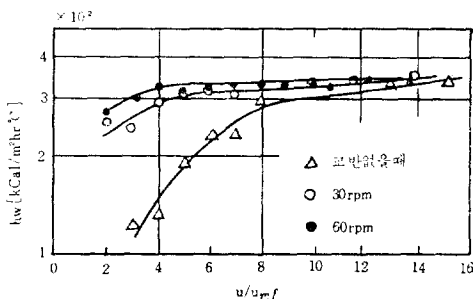


그림 1.  $h_w$  vs.  $U/U_{mf}^{(305)}$

#### b. 充填流動層

氣體流動層이 傳熱係數가 크고 層內에서의 粒子의 供給과 排出이 용이하다는 장점을 지니고 있지만 流動化 氣體의 대부분이 層內에서 氣泡로서 上昇하기 때문에 粒子와의 接觸이 나쁠 뿐만 아니라 氣泡徑 및 그 上昇速度가 流動層의 內徑과 分散板의 構造, 層高, 氣體流速, 粒子徑 및 그 密度등에 크게 영향을 받기 때문에 層內에서의 流體의 거동을 파악하기 어렵고 裝置를 擴大化하는데 곤란하다. 이런 缺點을 보완하고 장점만을 有効하게 利用하기 위해 層內에 여러 형태의 充填物을 넣어 그 빈틈에서 粒子가 流動化하여 氣泡의 合致成長을 막고, 粒子—流體間의 接觸을 원활하게 할 수 있을 뿐더러 層內에서의 粒子 및 流體의 흐름이 정상화되어 裝置의 擴大化가 용이하다.

또한 氣泡成長이 비교적 작기때문에 層內徑에 비하여 層高가 큰 경우에도 다른 流動層에서 보이는 Slugging 현상이 나타나지 않고 氣體流速의 증가와 더불어 均一하게 팽창한다. 充填物의 直徑은 層內의 粒子混合에 큰 영향을 주는데, 이것에 대해서는 半徑方向과 軸方向에 대해서 많은 研究가 報告되어 있다.

充填流動層內에서의 有效熱傳導度값은 固定層에 비해서는 500배 이상 크지만 일반 流動層에 비해서 작은 경향이 있으나 插入充填物의 直徑에 의해 그 값이 변한다. 熱傳達特性은 一般流動層과 유사한 경향을 가지며 동일粒子, 동일流動化氣體速度에서의 熱傳達係數는 一般流動層의 약 50~75% 정도의 값이 된다.

充填流動層에서도 一般流動層과 같이 氣泡相과 濃厚相이 存在하나 層內에서의 氣泡는 充填物에 의해 그 크기가 제약받기 때문에 氣體流速을 증가시키면 그數와 上昇速度만이 증가되어 一般流動層에서 사용되는 hydrodynamic flow model을 적용할 수 없어 流體—粒子間의 精確한 模型을 세우기 위해서는 대략 다음과 같은 假定을 하고 있다.

- i) 層內에서 氣泡相과 濃厚相이 存在한다.
- ii) 氣泡相은 plug flow한다.
- iii) 濃厚相에서의 氣體는 完全混合 또는 다른 여러조건(前報의 表참고)으로 간주한다.
- iv) 二相間에는 各 相內의 濃度差(즉分子擴散)에 의한 傳達과 相間交換係數(前報참고)로 표시되는 항에 따라 物質傳達이 일어난다.
- v) 層內에서의 氣泡體積은 層팽창에 의해 증가한 것과 같다고 본다.

이와 같은 充填流動層은 固定層과 流動層의 中間性質을 나타내는데 充填物의 크기와 形狀을 바꿈으로써 裝置의 特性을 살릴 수 있어 각종 工業에 應用되고 있다.

#### c. 攪拌流動層

反應速度가 느린物質을 流動層에서 合成, 酸化, 또는 還元하는 경우 傳換率이 低下되는데 이를 올리기 위해 氣體流速을 줄이면 정상적 流動化가 안되기 때문에 곤란하다. 이런 경우 氣體의 最小流動化速度 부근이나 그 以下에서 攪拌器를 부착시켜 攪拌하며 反應시킬 경우 보다 큰 傳換率을 얻을 수 있을 뿐더러 다음과 같은 利點을 갖고 있다.

- i) 多樣한 粒子의 觸媒를 使用할 수 있다.
- ii) 觸媒全部가 反應에 기여하기 때문에 (dead zone이 감소) 反應率이 증가한다.

- iii) 操作하는 도중에 觸媒交替가 용이하다.
- iv) 均一溫度 유지할 수 있다
- v) channeling을 방지하고 攪拌抵抗을 줄일 수 있다.

또한 吸濕性이 큰 부착성粉體나 處理도중 凝集되기 쉬운 粒粉體를 處理할 때도 마찬가지로 操業을 용이하게 하기 위해 기계적으로 粉粒子的 分散을 조장하는 攪拌器가 필요하다.

Kawabata<sup>251)</sup>의 技術報告에 보면 攪拌流動層을 이용하여 石炭을 乾留하는데 應用한 바 있다 그림 2에 표시한 바와 같이 流動化空氣中の 酸素의 未反應率  $\frac{C}{C_0}$ 와 空氣平均滯在時間  $\theta_g (= \frac{H_f}{u_0})$ 와의 관계를 나타냈는데 꽤 유리한 것으로 생각된다.

이와 같은 攪拌流動層에 의해 이제까지 연속 조업이 不可能하던 粉鐵鑛石의 還元, 粘結性物質의 乾燥, 石炭의 乾留, 廢棄發의 燒却등에 널리 應用되리라 본다.

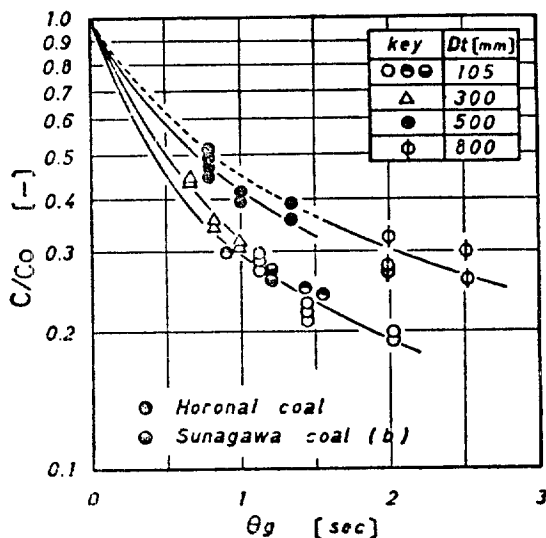


그림 2. Relationship between  $\frac{C}{C_0}$  vs.  $\theta_g$

#### d. 振動流動層

보통流動層 反應器에서 流動化 시키기 어려운 粒子를 다루는 경우 振動과 衝擊을 더해주면 流動化狀態가 向上되고, 最小流動化速度를 줄일

수 있으며 粒子—流體間의 熱 및 物質傳達를 증진시키며 層과 隔壁間의 傳熱도 향상시킬 수 있다. 다음 그림 3은 流動層에 振動을 가했을 때와 가하지 안했을 경우 氣體流速과 傳熱係數와의 관계를 나타내고 있는데 流動化 開始速度以下에서 이미 固定層보다 훨씬 큰 傳熱係數를 나타내고 있다.

또한 加振에 의해 加熱面 부근에 있는 粒子의 更新速度가 증가하여 流動이 활발해지고 層間差가 비교적 작기 때문에 加熱面에서의 粒子密度가 氣體流速을 크게 하더라도 감소하지 않아 振動強度의 증가에 의해 傳熱係數가 현저하게 커진다.

振動流動層乾燥器의 경우 드럼乾燥器性能과 비교해 보면 表 2에서 보는 바와 같이 그 성능이 월등함을 알 수 있다.

振動流動層에서는 기계적진동을 가할때 材料의 강도및 장치를 고려해야하는 문제점이 있으나 濕潤性粒子和 같은 難流動性 物質의 流動化를 촉진시키는데 利用할 수 있으며 加振에 의해서 粒子의 운동이 활발하기 때문에 乾燥를 목적으로 하는 경우 局部加熱을 막을 수 있다. 또한 層內에 脈動流가 생김으로서 粒子—流體間의 相

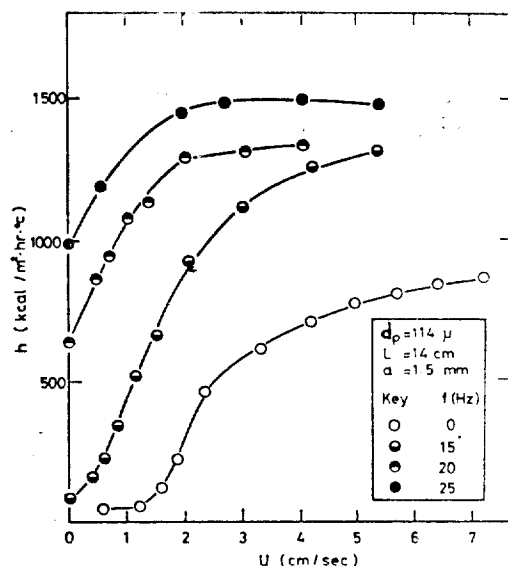


그림 3. Comparison of heat transfer coefficient in vibro-fluidized bed and in fluidized bed<sup>202)</sup>

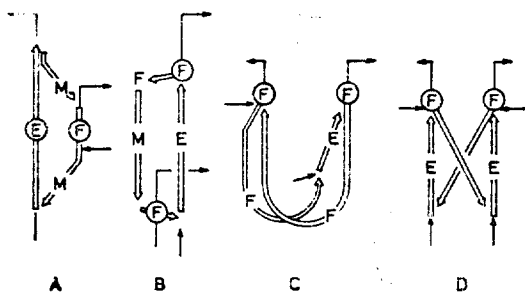
표 2. Comparative characteristics of drum type drier and vertical vibro-drier<sup>2)</sup>

| Characteristics                                     | Drier |          |
|---|-------|----------|
|   | Drum  | Vibro-FB |
| Drying time, min                                    | 20    | 4        |
| Electric power Consumption, KWH /ton                | 2.21  | 1.77     |
| Moisture removed from 1m <sup>2</sup> , kg/hr       | 0.61  | 2.66     |
| Specific heat consumption, kcal/kg H <sub>2</sub> O | 1500  | 850      |

對速度가 증가하기 때문에 傳熱 및 物質傳達速度가 증가하는 利點이 있다.

### e. 循環流動層

최근 석유자원의 有効使用과 完전 脫黃이 사회적인 관심사가 되고 있는데 이런 문제를 완전히 해결하기 위한 시도로서 각종 原油, 殘渣油 및 重質油의 熱分解와 氣體化에 대한 연구가 進行되고 있다. 이에 적합하고 유리한 것중의 하나가 循環流動層이다. 이는 대개 二室流動層을 적당히 조합하여 한 系를 이루는데, 이系內에는 移動層(M), 流動層(F), 그리고 氣流輸送層(E)部分이 있게 된다. 그림 4에 기존 工業적 循環系를 간단히 표시했다. 重質油를 熱分解하여 Olefins類를 제조한다고 생각 할때 反應時間이 짧



A: Lurgi's sand cracker

B: UOP's FCC unit

C: ERE model IV FCC unit

D: BASF fluidized flow process

White circles mean the main reacting zones

그림 4. Schematic diagram for various circulation<sup>146)</sup> systems in practical application

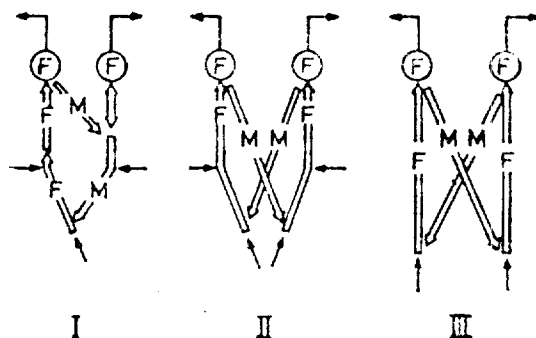


그림 5. Schematic diagram of circulation systems

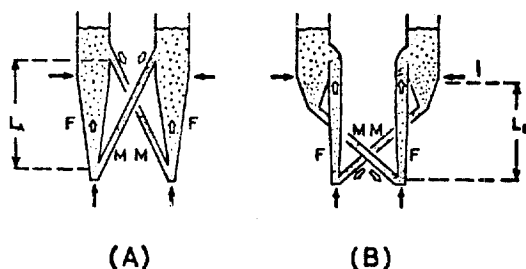


그림 6. Two possible systems for No. III in Fig. 5.

아야하고 生成氣體를 急冷해야 하는데 이럴경우 이 循環流動層系가 비교적 적합함을 알 수 있다

Kunii<sup>146)</sup>가 소개한 流動層-移動層을 조합한 三個의 系를 그림 5에 표시했다. 그리고 그림 6는 그림 5에 표시한 III형 循環系를 나타냈다.

固體循環系는 이온교환수지 재생, 觸媒의 재생, 熱交換器等 그 應用이 다양하리라 생각된다

## 7. 結 言

流動層反應器를 고려할때 分散板設計, 內插物形狀의 效果, 流動粒子和 壁面間摩耗, 高溫流動層에서의 濕度和 粒子間引力 등 해결하여야 할 과제가 많은 것이다. 이를 위하여 模型을 기초로 어느정도의 流動層反應器 履歷을 파악하고 좀더 계통적실험에 의한 신뢰할만한 實測值가 集積되어져야만 하겠다.

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