IAEA -SM-350/38

XA9847740

APPLICATION OF RADIOISOTOPE TRACER TECHNIQUES IN EVALUATION OF IRRADIATION VESSEL OF FLUE GAS TREATMENT SYSTEM

JOON-HA JIN, MYUN-JOO LEE,
SUNG-HEE JUNG, YOUNG-CHANG NHO
Korea Atomic Energy Research Institute,
Taejon, Republic of Korea

Abstract

The proper design of the irradiation vessel of electron beam flue gases treatment plant and resultant optimum gas flow pattern is a very important factor to a get high removal efficiency of toxic materials from flue gases. Radioisotope tracer experiments were conducted to study the residence time distribution of gas flow in a cylindrical irradiation vessel. A few mCi of gaseous radioisotope tracer Ar-41 was injected to the upstream of the vessel and the input and output response were measured with two NaI scintillation detectors. The same experiment was conducted after the modification of the vessel by introducing 4 baffles.

The experimental data were analyzed to calculate mean residence times and mixing characteristics of each system using the residence time distribution (RTD) analysis software. A method to estimate pollutant removal efficiencies of an irradiation vessel from the residence time distributions measured by radiotracer experiments was suggested. The analytical results were compared to evaluate the effect of the baffles on the removal efficiency of the plant.

1. INTRODUCTION

A new technology has been developed to purify flue gases from coal burning boilers and incinerators using electron beam accelerators [1]. The Electron Beam Process is a dry-scrubbing process which simultaneously removes sulfur dioxide (SO_2) and nitrous oxides (NO_x) from combustion flue gases. The irradiation of the flue gas produces active radicals and atoms which react with the SO_2 and NO_x to form their respective acids. In the presence of ammonia (NH_3), these acids are converted to ammonium sulfate nitrate ($(NH_4)2SO_4 \cdot 2NH_4NO_3$) which can be used as fertilizer.

Even though it has a long history of various researches and many technical advantages to conventional processes, the industrialization of the process has not been fast enough. The improvement of its economic competitiveness to conventional processes with higher removal efficiency of toxic materials from flue gases will encourage the industrialization of the new process. The appropriate design of the irradiation vessel is one of the most important factors to get the maximum removal efficiency and the economic competitiveness.

All the process vessels are designed to carry out a specific function. When the vessel fails to perform this function there may be either a design fault or a malfunction of some sort, and residence time distribution (RTD) analysis is often used to gain an understanding of what is happening inside the vessel.

Tracer technology is being used in many diverse scientific disciplines to help gain a better understanding of dynamic processes. Radioactive materials are particularly effective as tracer in process investigations of vessel systems because of the wide variety of isotopes available, chemical and fluid compatibility, the low concentration required, and the sharp pulse of tracer may be injected into most systems. The ability to detect the movement of tracer from outside of vessel with on line method is another important advantage of the radiotracer technology [2].

To gain knowledge for the design of the irradiation vessel, a series of radiotracer experiments were conducted using Ar-41 as tracer. The experimental data were analyzed to calculate mean residence times and mixing characteristics of the system using the residence time distribution analysis software which was developed by Korean tracer group. A method to estimate pollutant removal efficiencies of the irradiation vessel from the residence time distribution measurements is suggested. The analytical results were compared to evaluate the effect of the baffles on the removal efficiency of the plant.

2. IRRADIATION VESSEL

The irradiation vessel in which chemical reactions are initiated by high energy electron beam is the most important part of the flue gas treatment system because the biggest portion of electric energy is consumed in this process. The vessel of the pilot scale electron beam flue gas treatment system is a cylindrical stainless steel vessel with thin titanium window on the top to accept electron beam. The diameter of the vessel is 27cm and the length is 118cm and the volume of the vessel is divided to 5 sections with 4 baffles.

3. RADIOTRACER

Among gamma emitting gaseous radioisotopes, Ar-41 was employed as tracer because the atomic weight of the isotope is most similar to those of the main components of flue gases. It emits 1.29MeV gamma-ray and has a short half life of 110 minutes. It was produced by the neutron irradiation of argon gas sealed in a quartz ampoule.

A radiotracer injection system was designed as shown in Fig. 1 to get sharp input impulses and to minimize exposure of operators to the radiation. The operation procedure is as follows;

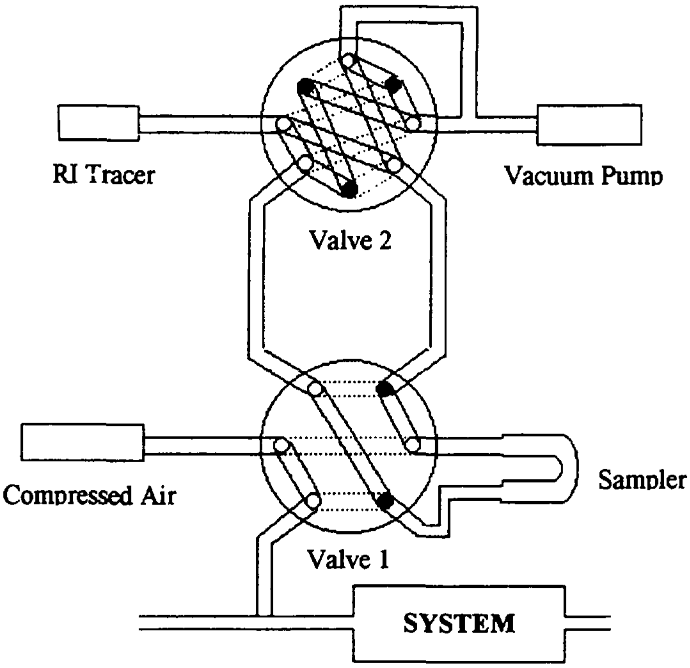


Fig.1. Gas tracer injection system

1) Connect the ampoule containing Ar-41 to the injection system through Tygon tube.
2) Evacuate the Sampler and the line up to the Tygon tube with the vacuum pump.
3) After turn Valve 2 to charge position, break the tip of the quartz ampoule in the Tygon tube to fill the tracer gas in the Sampler.
4) Turn Valve 1 to discharge position to inject the tracer gas to the system.
5) For the preparation of the second injection, evacuate the sampler but not the line to RI tracer ampoule.
6) Repeat step 3), 4) and 5) for the second injection.

4. EXPERIMENT

The first experiment was conducted with the vessel without the baffles. Two NaI scintillation detectors (2 inch in diameter and 2 inch in length) were installed on inlet and outlet pipe of the vessel with lead collimators and were connected to the rate meters (Eberline Model ESP-2) as shown in Fig. 2. The signal from the rate meters were introduced to a data acquisition system to log the data simultaneously.

After turn on the rate meters and data acquisition system, a few mCi of gaseous radioisotope tracer Ar-41 was injected to the inlet pipeline through the injection point installed at approximately 10m upstream of the pipeline from the inlet detector. As the diameter of the pipeline is only 1 inch, the mixing length(10m) was far more than enough. The counts from the two detectors were logged every 2 seconds.

The second experiment was conducted with the same operating condition after install 4 baffles in the vessel.

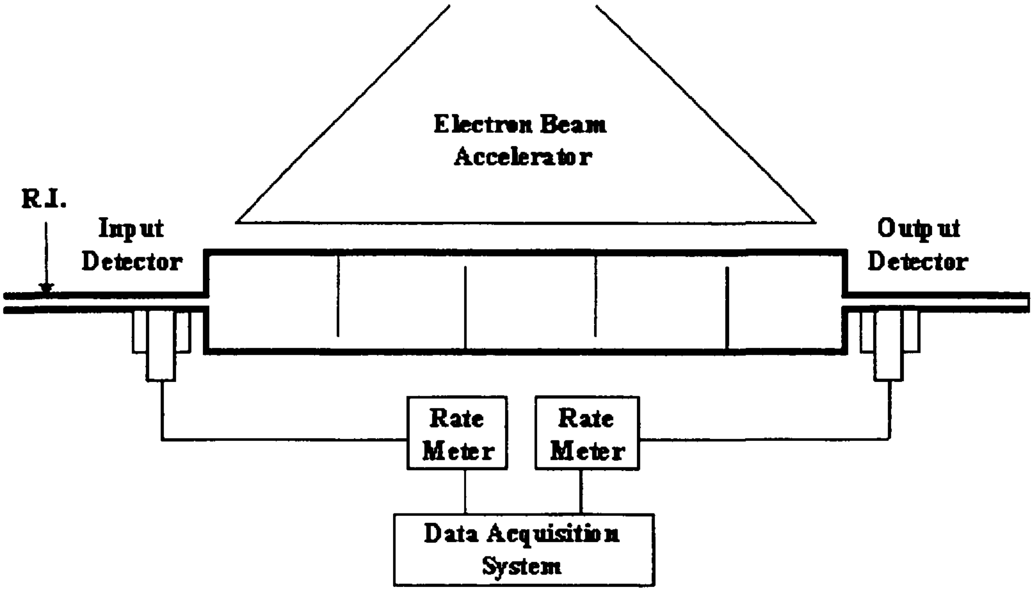


Fig. 2. Arrangement for RTD study using radioisotope as tracer

5. RESULTS AND RESIDENCE TIME ANALYSIS

The experimental data were analyzed with the RTD Analysis Program which had been developed by KAERI tracer team. The input and output response for the two experiment are shown in Fig. 3-a and Fig. 3-b together with simulated output responses(dotted line) obtained by the Perfect Mixers in Series Model.

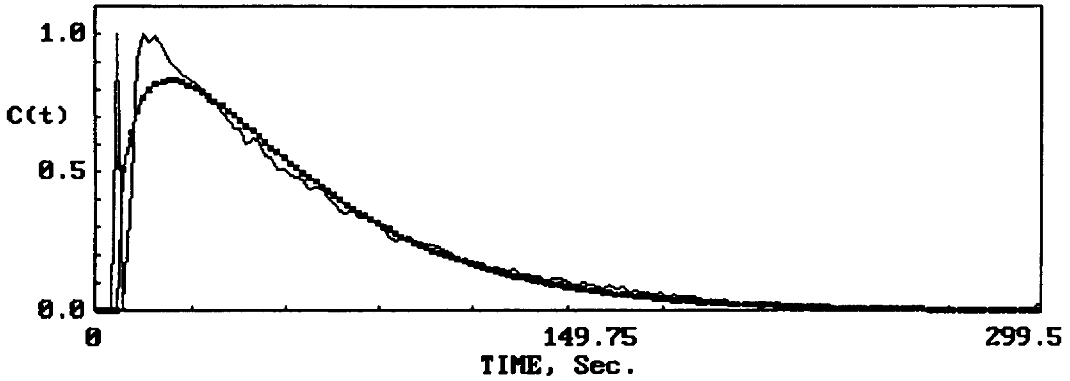
The mean residence times (MRT) of the first (without baffles) and second (with baffles) experiment are calculated as 56.9 sec. and 63.8 sec., respectively. It is not clear whether the difference of MRT came from the flow rate change or dead volume of first system.

The shapes of the output peaks are clearly different to each other. The peak for the vessel without baffles is similar to the response function of a perfect mixer, and fit with the simulated response function with tank number 1.4. On the other hand, the peak for the vessel with 4 baffles fit well with the simulated response function with tank number 3.9. This result means that there are quit a big portion of by-passing and back mixing of flue gases in the first system, but these undesirable phenomena can be reduced by installing baffles in the vessel.

6. EFFICIENCY ESTIMATION

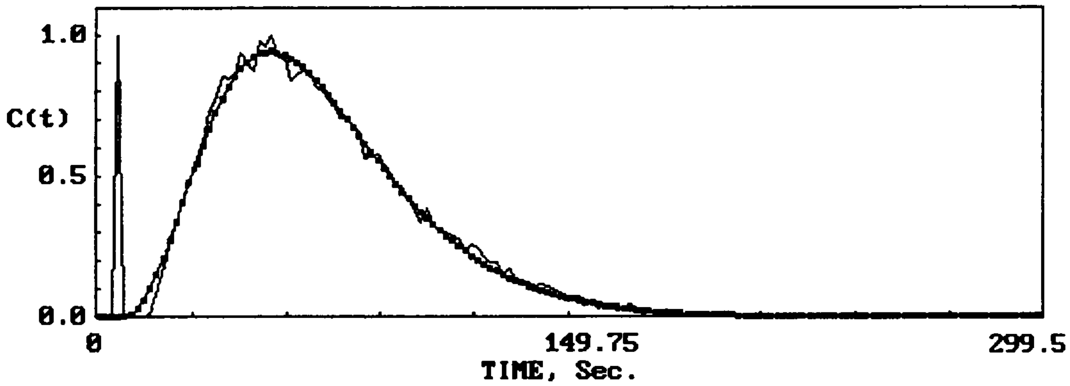
The impulse response function of a system with MRT τ and tank number N is as follow [3];

$$H(t) = (\frac{N}{\tau})^N \frac{t^{N-1}\exp(-\frac{N}{\tau}t)}{(N-1)!}$$



```
MEAN RES. TIME =  56.91 Sec.    @  7.02 →  63.93 (  63.62 :SIM.)
TANK NUMBER    = 1.4            @  RMS (A-NOMALIZATION) : 0.058
```

Fig. 3-a. RTD of flue gas in the cylinderical irradiation vessel with no baffle



```
MEAN RES. TIME =  63.82 Sec.    @  7.04 →  70.86 (  70.86 :SIM.)
TANK NUMBER    = 3.9            @  RMS (A-NOMALIZATION) : 0.022
```

Fig. 3-b. RTD of flue gas in the cylinderical irradiation vessel with 4 baffles

The impulse response functions of the two systems can be obtained by substitution of the two parameters (MRTs and tank numbers) measured by the tracer experiments (see Fig. 3-a and Fig. 3-b) to the above equation.

$H_a(t) \ = \ 6.42 \times 10^{-3}\, t^{0.4}\, e^{-0.0246t}$ (for the vessel without baffles)

$H_a(t) \ = \ 3.46 \times 10^{-6}\, t^{2.9}\, e^{-0.061t}$ (for the vessel with 4 baffles)

On the one hand NO_x is decomposed by radiation, on the other hand it is produced by the radiolysis of air. Thus, the change of NO_x concentration with radiation dose(D) can be presented as follow;

$$\frac{d[NO_x]}{dD} \ = \ -k[NO_x] + k'[N_2][O_2]$$

As k' and the concentration of air components in flue gas can be considered as constant, the second term of the equation can be replaced with constant "a". Then,

$$\frac{d[NO_x]}{a - k[NO_x]} \ = \ dD$$

The solution of this differential equation and the removal efficiency E(D) are as follow;

$$[NO_x] = \frac{a}{k}(1-e^{-kD}) + [NO_x]_0 \, e^{-kD}$$

$$E(D) = \frac{[NO_x]_0 - [NO_x]}{[NO_x]_0} = (1 - \frac{a}{k[NO_x]_0})\,(1-e^{-kD})$$

As a, k and $[NO_x]_0$ are constants, the first parenthesis of the equation can be substituted with constant "C" as follow;

$$E(D) = C\,(1-e^{-kD})$$

The experimental NO_x removal efficiency vs. radiation dose was reported as shown in Fig. 4 [4]. The equation representing the curve is obtained by fitting method with Sigma Plot program as follow;

$$E(D) = 0.77\,(1-e^{-0.2069D})$$

where E(D) is the NO_x removal efficiency with radiation dose of D kGy. As the dose is the product of dose rate and time (D=Dr ×t), the equation can be written as follow;

$$E(t) = 0.77\,(1-e^{-0.2069Dr\cdot t)})$$

As the H(t) function is area normalized, H(t)×E(t) is the removed portion of NO_x from the flue gas which remain in the vessel for time t, and $\int_0^\infty H(t)\cdot E(t)\,dt$ is total NO_x

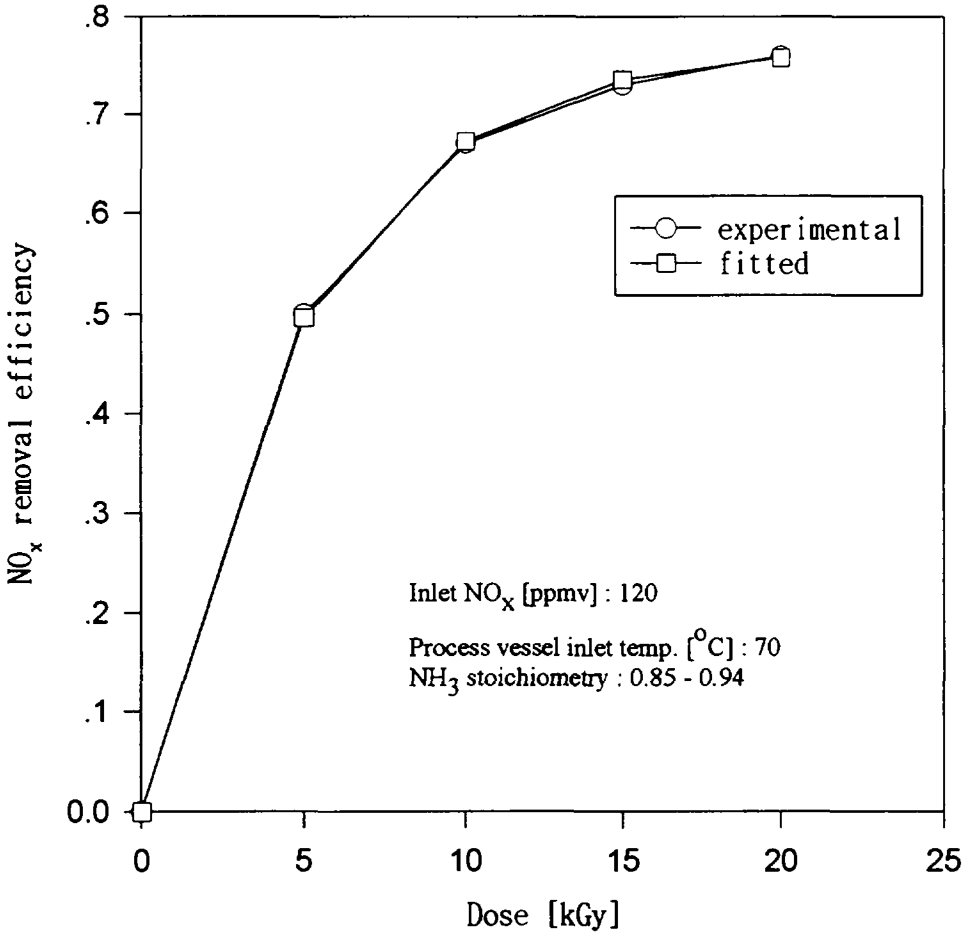


Fig. 4. Experimental and fitted curve of NO_x removal vs. Dose

removal efficiency of the vessel at a dose rate of Dr. The ratio of the area under H(t)×E(t) curve in Fig. 5 to the area under H(t) curve is the removal efficiency at the dose rate of 0.2 kGy/s and appears to be 67%.

Figure 6 shows some examples at different dose rates assuming that the gas flow rate is fixed. It is appeared that the removal efficiencies of the vessel with baffles are higher than those without baffles especially at low dose rate. The efficiencies of the two systems at different dose rates are given in Fig. 7. The efficiency of the vessel with baffles reaches 97 percent of saturated value(maximum removal efficiency, 77%) at the dose rate of 0.4 kGy/sec, whereas 87 percent for the vessel without baffles. The figure shows that the installation of baffles are effective in increasing removal efficiency especially in low dose rate.

Even with the vessel without baffles the maximum removal efficiency 77% can be reached at the dose rate higher than 5 kGy/s. In this case, however, the energy efficiency of the system will be very low, and the process can not compete with conventional processes. Thus, an optimum removal efficiency should be selected by considering both economics and environment. If 67% (87 percent of the maximum removal efficiency) is selected, the dose rate of 0.4 kGy/s should be employed with the

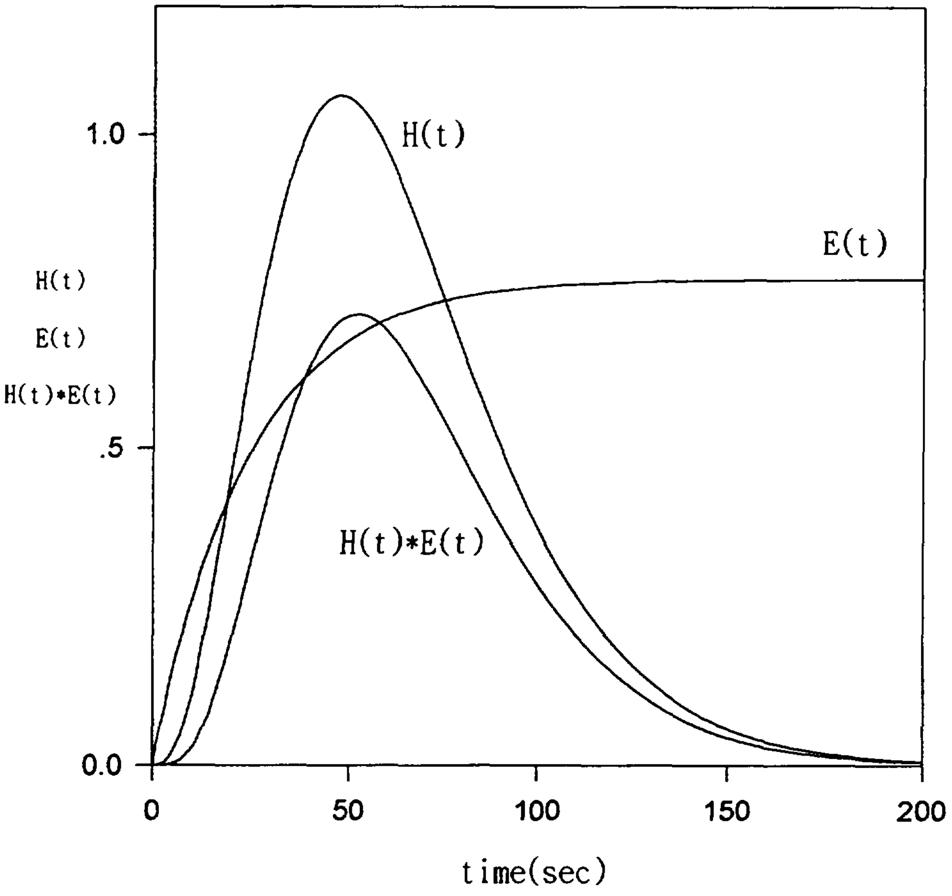


Fig. 5. NO_x removal efficiency at 0.2kGy/sec

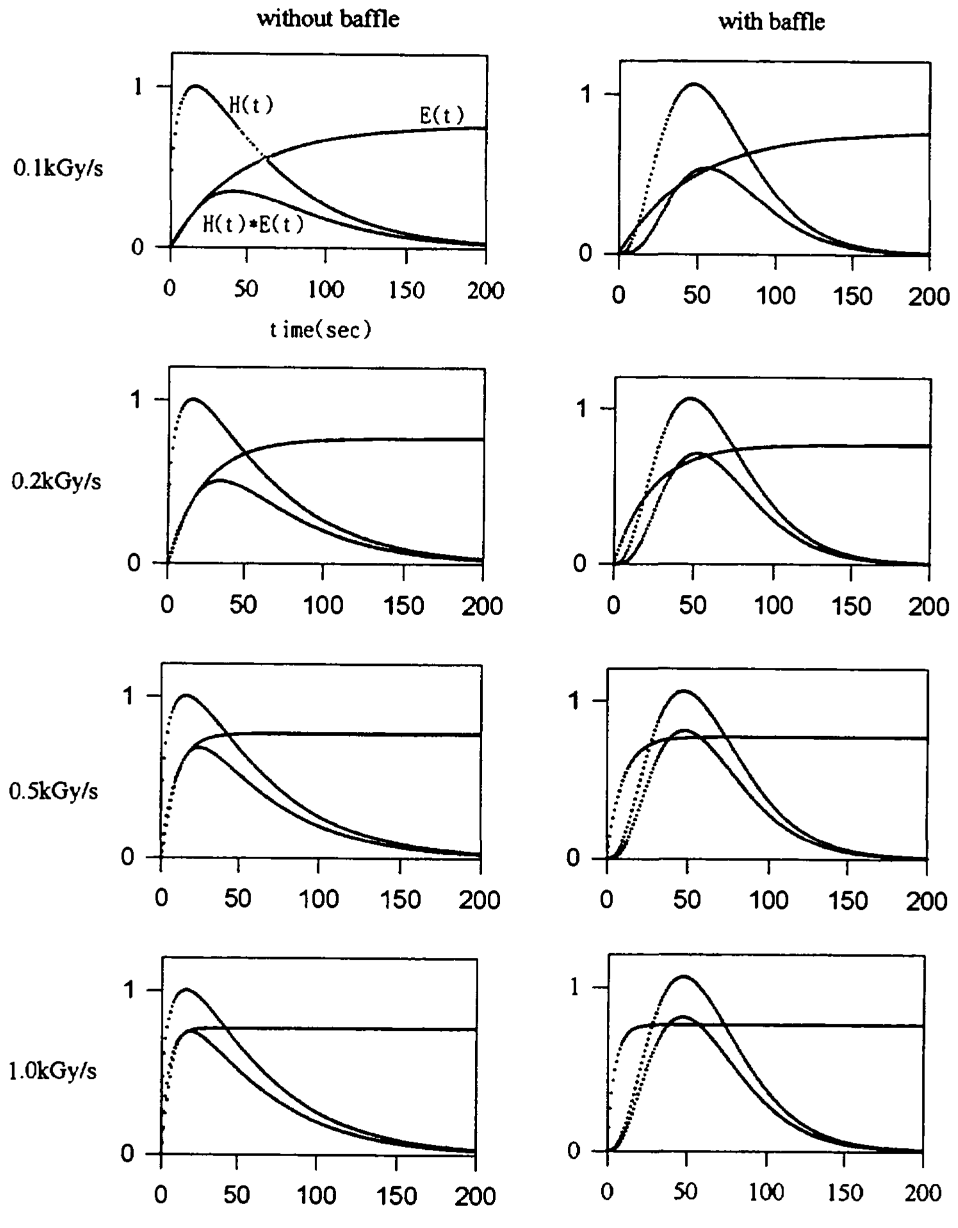


Fig. 6. NO_x removal efficiency vs. dose rate

vessel without baffles, whereas only 0.2 kGy/s is enough with the vessel with baffles to get the same removal efficiency. This means that 50% of electric energy can be saved simply by employing 4 baffles in the irradiation vessel. As another example, dose rates of 1.5 and 0.5 kGy/s should be employed to remove 75% of NO_x with the vessels without and with baffles, respectively. It means that the same removal efficiency can be achieved with one third of energy by employing the baffles in the vessel.

The efficiency estimation method suggested in this paper may have some errors, because other factors, such as uneven distribution of dose rate in the vessel and lose of electron beam by the metal baffles, are not considered. It was intended to show the way how to use tracer techniques for the design of the vessel to get better NO_x removal efficiency.

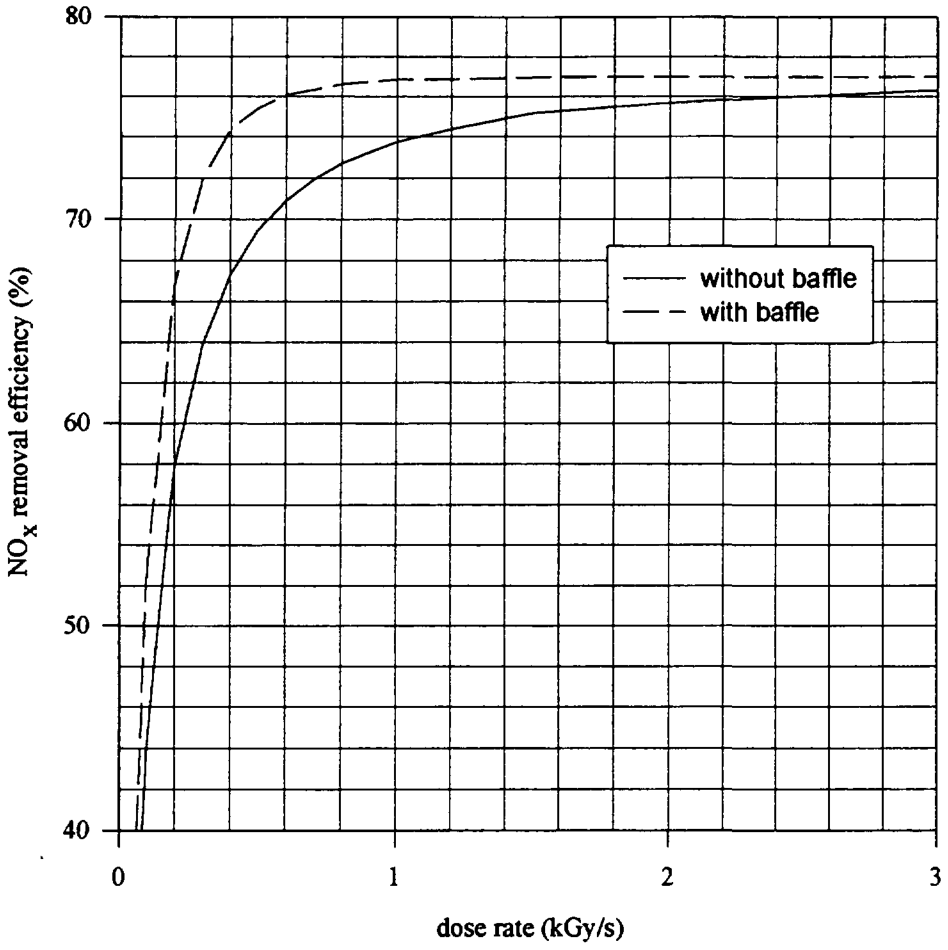


Fig. 7. Effect of baffles on NO_x removal efficiency

REFERENCES

[1] KAWAMURA, K., AOKI, S., J. Atomic Energy Soc. Japan, 14, (1972) 597; MACHI, S., Proc. the 11th Japan Conference on Radioisotopes, (1973) 213

[2] CHARLTON, J. S., Radioisotope Techniques for Problem Solving in Industrial Process Plant, Leonard Hill, London, (1986) 112

[3] IAEA, Guidebook on Radioisotope Tracers in Industry, IAEA technical report series No. 316, (1990) 63

[4] CHMIELEWSKI, A. G., TYMINSKI, B., LICKI, J., ILLER, E., ZIMEK, Z., DOBROWSKI, A., Pilot plant for EB flue gas treatment - start up and two stage irradiation tests, Radiat. Phys. Chem. 42(4-6), (1993) 663