Transactions of the Korean Nuclear Society Spring Meeting
Jeju, Korea, May 18-19, 2023

Ethanol Effect in Soil Decontamination Process Using Supercritical Carbon Dioxide

Seungil Ha [a], Kwangheon Park [a*], Jinhyun Sung [a]
[a]*Department of Nuclear Engineering, Kyunghee University, Kyunggi-do, 446-701, Korea*
[*]*Corresponding author: kpark@khu.ac.kr*

1. Introduction

Decontamination of radioactive soil is difficult due to radionuclides being adsorbed into the pores of the soil. Recently, there has been an increasing interest in site remediation along with decommissioning, but current soil decontamination technologies have disadvantage of generating large amounts of secondary waste.

Soil decontamination technology using supercritical carbon dioxide($SCCO_2$) is being studied as an eco-friendly technology that fundamentally suppresses the generation of secondary waste. In this study, we confirmed the effect of ethanol on the decontamination efficiency and its mechanism in soil decontamination process using $SCCO_2$.

2. Methods and Results

2.1 Main Solvent: $SCCO_2$

The high penetration power of CO_2 is advantageous for decontamination of porous materials such as soil. CO_2 becomes supercritical state when it has a temperature (31.1 ℃) and pressure (73.8 bar) above the critical point, and it has the properties of liquid and gas at the same time. The diffusivity of the gas maximizes the penetration of CO_2, and the solubility of the liquid can soluble nuclides in the solvent penetrated into the pores. After decontamination, the solvent and the waste are separated through decompression to minimize the generation of secondary waste and to reuse the separated solvent.

2.2 Additives: Chelate Ligand and Co-Ligand

CO_2 is a nonpolar solvent, but most radioactive contaminants exist in polarity within the soil. Hence, the addition of chelate ligands which can extract polar contaminants and be soluble to $SCCO_2$ are needed for soil decontamination. In addition, during the extraction process, the efficiency decreases due to anions which are not soluble in $SCCO_2$. To compensate for this, the co-ligand was introduced as an additive. The co-ligand instead provides an anion which is well soluble in $SCCO_2$. Through previous studies, Catechol Amine was selected as the chelate ligand and NEt_4PFOSA was selected as the co-ligand [3].

2.3 Introduction of Co-Solvent

When constructing the decontamination process, the additive is more efficient in a liquid phase than in a solid phase. In this study, ethanol was introduced as co-solvent for liquid additives and efficiency improvement. Ethanol can completely dissolve the additive and is simultaneously soluble in $SCCO_2$. In addition, since ethanol has its own decontamination effect, it can serve as co-solvent in $SCCO_2$ decontamination process. In this study, the solubility of ethanol for $SCCO_2$ was measured, and the efficiency improvement due to the addition of ethanol in the decontamination process was confirmed.

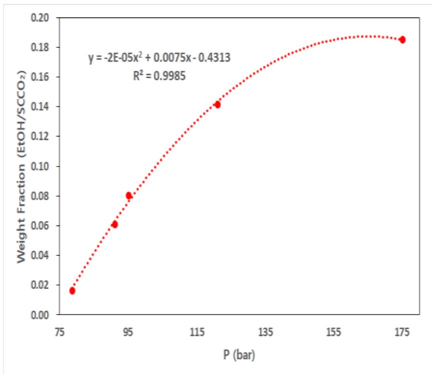

Fig. 1. The solubility of ethanol in $SCCO_2$

2.4 Sample Preparation and Decontamination

As for the soil specimen, chemically treated Sea Sand was purchased as a standard specimen. In addition, real soil was collected in the site of Kyunghee University and classified into coarse (0.5~1.0 mm), medium (0.25~0.5 mm), and fine (~0.25mm) according to the particle size. Simulated contaminated soil were prepared through adsorption using non-radioactive Cs, Sr, and U standard solutions on standard and real specimens.

The decontamination experiment was carried out for 90 minutes under 40 ℃, 200 bar, and an ultrasonic condition of 40 kHz, considering the critical conditions of CO_2 and the solubility of the additives. To confirm the decontamination effect, the concentration of nuclides in the soil before and after decontamination was measured through ICP-MS (Inductively Coupled Plasma-Mass Spectrometry). Thereafter, the decontamination efficiency was evaluated through the measured concentration of nuclides before and after decontamination.

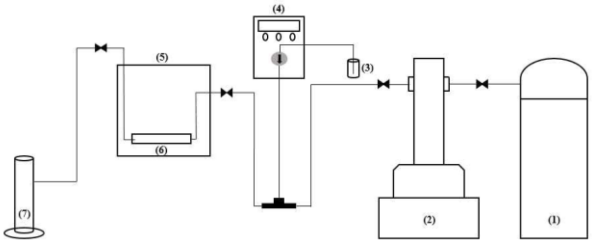

Fig. 2. Experimental apparatus for soil decontamination
(1) CO_2 Tank, (2) Syringe Pump, (3) Mixing cell, (4) High Pressure Piston Pump, (5) Ultrasonic wave Controller, (6) Reaction cell, (7) Bubbler

2.5 Ethanol Effect in Decontamination Process

The decontamination experiment to confirm the ethanol effect was conducted for each particle size. In the experiment, $SCCO_2$ and additives were sequentially added under a single ethanol decontamination condition. Also, the pretreatment effect was confirmed by adding a pretreatment process using ethanol before and after decontamination. As a result, it was confirmed that the decontamination effect of Cs was significantly lower than that of Sr under a single ethanol condition. However, as a result of using ethanol as co-solvent in the $SCCO_2$ process, the decontamination effect of Cs gradually increased and further increased after the use of the additives. Finally, as a result of ethanol pretreatment before and after decontamination, decontamination efficiency of about 80-90 % for each particle size was confirmed.

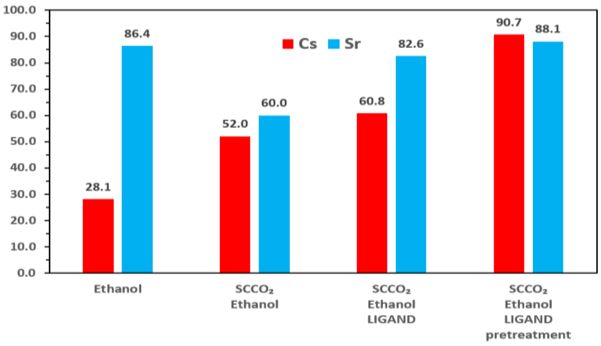

Fig. 3. Ethanol effect in soil decontamination (Coarse)

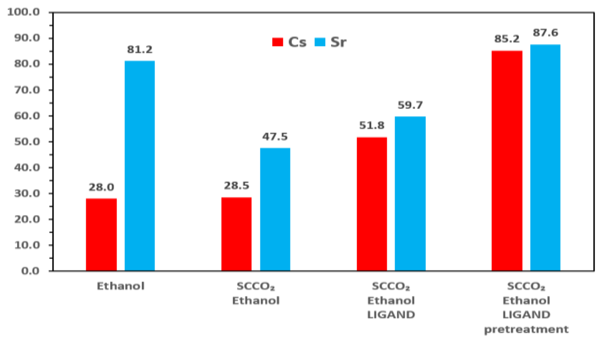

Fig. 4. Ethanol effect in soil decontamination (Medium)

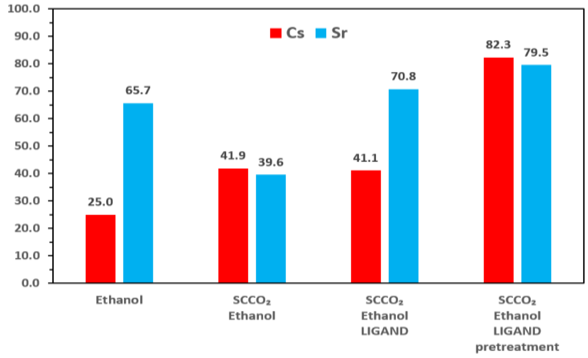

Fig. 5. Ethanol effect in soil decontamination (Fine)

3. Conclusions

This study aims to confirm the ethanol effect as co-solvent in the soil decontamination process using $SCCO_2$. Although there is a difference in decontamination effect depending on the adsorption characteristics of Cs and Sr in the soil, when additives and ethanol were dissolved in $SCCO_2$, both nuclides showed a high decontamination effect of about 80-90 %. In addition to this study, when the process volume increases for commercialization or the waste post-treatment process is added, it is expected that an eco-friendly soil decontamination process which minimizes secondary waste can be developed.

REFERENCES

[1] J. McHardy, S. P. Sawan, 1998, Supercritical Fluid Cleaning – Fundamentals, Technology and Applications, 1st ed, Noyes Publications.
[2] C. M. Wai, S. Wang, 1997, Supercritical fluid extraction: metals as complexes, J. Chromatography A, Vol.785, No.1 pp.369-383.
[3] K.Park, T.Kim, J.Park, X.Yan, H.Kim, 2020, Development of a carbamate-conjugated catechol ligand and its application to Cs extraction from contaminated soil by using supercritical CO2, Chemosphere 242(2020), 125210.
[4] T. Suzuki, N. Tsuge, K. Nagahama, 1991, Solubilities of ethanol, l-propanol, 2-propanol and lbutanol in supercritical carbon dioxide at 313 K and 333 K, Fluid Phase Equilibria, 67(1991), pp.213-226.