# *In Vitro* Gastric and Intestinal Bioaccessibility of Cesium from Ingested Contaminated Concrete Waste

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## 1. Introduction

Cesium-137 ($^{137}$Cs) is a man-made radionuclide that can be produced as a by-product of nuclear power plant operations or accidents. Exposure to these substances can pose a significant risk to human health. Inadvertent ingestion of soil, dust, or concrete containing $^{137}$Cs, through inhalation or hand-to-mouth activities, can result in an increased internal radiation dose. $^{137}$Cs are absorbed through oral exposure or inhalation and are distributed uniformly in the body. It competes with potassium (K) for membrane transport [1]. The bioavailable fraction of $^{137}$Cs is defined as the portion of these materials that reaches the systemic circulation in the human gastrointestinal (GI) tract [2]. Common animal models, such as rats and mice, have been used to determine the bioavailability of contaminants in soil for human health exposure assessment [3]. However, these tests can be expensive and time-consuming. Therefore, several *in vitro* assays have been developed, including the Unified BARGE Method (UBM), Physiologically Based Extraction Tests (PBET), and Solubility Bioaccessibility Research Consortium (SBRBC) to simulate the human GI condition which includes gastric (GP) and intestinal phases (IP) [2]. In this study, the amount of dissolved Cs by the extraction from *in vitro* GI fluids is measured and referred to as bioaccessible. This represents the maximum amount of contaminant that can potentially be absorbed by the intestines and transferred to the bloodstream. Herein, this study aims to assess the bioaccessibility ($f_{ba}$) of Cs using the UBM method in Cs-contaminated concrete (CC). The research outcomes will contribute significantly to enhancing our knowledge of Cs exposure and ingestion risks, paving the way for better risk management strategy to the radiological workers in the nuclear facilities.

## 2. Materials and methods

### *2.1. Concrete preparation*

Preparation of Cs-contaminated concrete (CC), using non-radioactive forms of Cs, followed the methodological guidelines provided by Han et al. [4]. To guarantee the homogeneity of the concrete composition, a precisely measured mixture of 14.0 g of Portland cement (OPC, Type I), 3.4 g of fly ash, and 24.6 g of dry sand were thoroughly mixed by hand for 5 minutes. After that, 8.0 mL of a well-prepared 100 ppm solution of surrogated stable Cs was carefully added to the concrete slurries. The concrete slurry was cured in a cylinder-shaped paper mold in a sealed container with water at the bottom over 28 days. Post-curing, the CC samples were pulverized and filtered to a particle size of <75 μm. Concrete particles less than 75 μm were selected under the assumption of ease of finger adherence and a high probability of being ingested incidentally [5].

### *2.2. UBM in vitro assay*

The bioaccessibility ($f_{ba}$) of Cs in CC was analyzed using the UBM *in vitro* assay to mimic the human digestive processes. This study modified the procedure to 0.2 g with 20 mL UBM fluid (Figure 1). The standard UBM *in vitro* procedure is available in the literature [6]. Following the completion of *in vitro* assays, GP and IP samples were centrifuged at 3000 rpm and filtered through 0.45 μm before analysis by an inductively coupled plasma mass spectrometer (ICP-MS). All GP and IP extractions were performed in triplicate for each soil sample. The Cs-$f_{ba}$ was computed and expressed as follows:

$$(1)\quad f_{ba}\,(\%) = \frac{\text{bioaccessible concentration (mg kg}-1)}{\text{total concentration (mg kg}-1)} \text{x}100$$

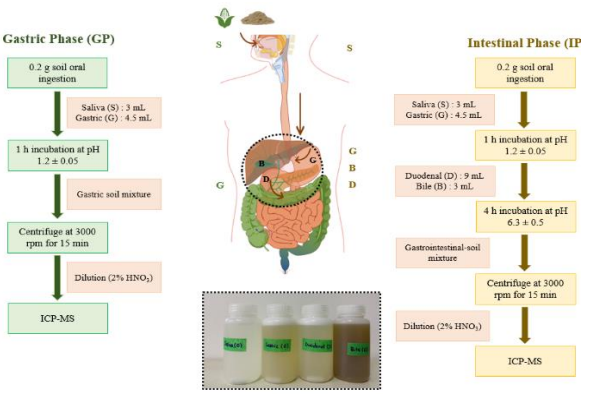

Fig. 1. Schematic diagram of UBM *in vitro* assays [6].

## 3. Results and discussion

### *3.1. Cs-contaminated concrete*

The instrumental neutron activation analysis (INAA) method was employed to determine the total concentration of Cs present in the contaminated concrete. The results showed that the concentration of Cs in CC was 106.62 ± 1.40 mg $kg^{-1}$, and Cs spike recoveries were ranged from 84–124%. These findings not only confirm that Cs is indeed incorporated within the concrete, but also demonstrates that the material is suitable for further bioaccessibility studies. The X-ray diffraction (XRD) patterns of the simplified artificial systems are reported in Fig. 2(A), showing the main mineralogical phases detected. The XRD results reveal a major diffraction pattern that exhibits three main phases: quartz ($SiO_2$), calcite ($CaCO_3$), and ettringite ($Ca_6Al_2(SO_4)_3(OH)_{12}\cdot 26(H_2O)$). Analysis through XRD did not detect any Cs containing crystalline phases in the CC samples, possibly due to their low concentration below the detection limit. The major diffraction pattern is consistent with the Field Emission Scanning Electron Microscopes (FE-SEM) images, which depict a homogenous distribution of Si, Al, Ca, Fe, O, and S throughout the CC samples.

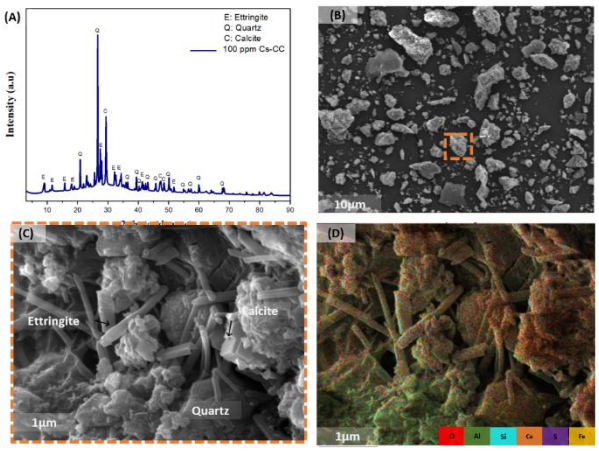


Fig. 2. XRD pattern (A) and FE-SEM-EDS image of Cs-contaminated concrete (B-D).

### *3.2. Assessment of Cs bioaccessibility*

According to Figure 3, the bioaccessibility of Cs in the GP assay ranges from 61.77% to 84.96% (with an average of 72.24%), while in the IP assay, it ranges from 57.38% to 61.82% (with an average of 60.76%). These empirical observations suggest a predominant bioaccessibility of Cs in the GP phase compared to the IP phase. It is pertinent to underscore that Cs contained within concrete (CC) is soluble under acidic conditions, given that the pH of the GP phase (pH 1.2) significantly surpasses that of the IP phase (pH 6.5). Such acidic environments foster reactions that considerably facilitate the release of Cs from the concrete matrix, particularly for Cs compounds such as cesium hydroxide (CsOH), which exhibit high solubility in acidic conditions, facilitating the dissociation of $Cs^+$ ions into the solution. Additionally, Kaminski's research reveals that quartz and calcite exhibit minimal affinity for Cs, potentially contributing to the facile solubilization of Cs, especially within acidic contexts [7]. Notably, iron and quartz have been identified as compounds capable of exerting a notable influence on Cs bioaccessibility, with their presence correlating to increased Cs desorption from the concrete matrix [7].

As the pH level increases, the bioaccessibility of Cs in IP decreases in comparison to GP. We have previously observed that Cs are highly mobile as $Cs^+$ in low-pH environments. However, in high pH conditions, certain phases within the concrete matrix (such as calcium silicate and calcium hydroxide) can be dissolved, which may cause the $Cs^+$ bound within these phases to be released into the solution [8]. $Cs^+$ can also substitute the cations present in the IP by replacing with $K^+$, $Ca^{2+}$, and/or $Na^+$, making it more bioaccessible [8]. The extent of bioaccessibility of $Cs^+$ from CC can vary based on the mineral phases present in the concrete, the pH of the gut fluids, ion exchange, and the impact of the concrete matrix on $Cs^+$ mobility in different pH environments.

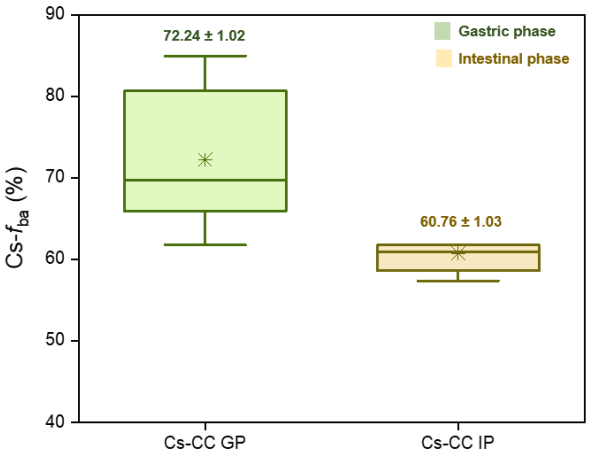


Fig. 3. The bioaccessibility of Cs in the GP and IP after oral exposure of Cs-CC.

## 4. Conclusion

Several factors greatly influence the outcome of *in vitro* measurements. Some of these crucial factors include the composition and concentration of soil and solid materials, the pH medium, and ion-exchange in the digestive assay. Therefore, accurate measurement of these factors is essential to obtain reliable and meaningful results in *in vitro* tests. To gain a better understanding, additional experiments with variations in concentration or method should be conducted in the future.

## 5. Acknowledgments

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