Investigation of cross-sectional gas-solid distributions in spouted beds using advanced non-invasive gamma-ray computed tomography (CT)

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Abstract

The successful operation and safety of the Very-High-Temperature Nuclear Reactors (VHTR) extremely depend on the quality of the TRISO nuclear fuel coated particles. Hence, the fuel coating technology of TRISO particles, based on chemical vapour deposition (CVD) process, performed in gas-solid spouted beds, is of utmost importance. However, the deposition of the coating layers surrounding the kernel is a delicate process, and impacted by the hydrodynamics of spouted beds. Therefore, in this work, we have applied an advanced non-invasive gamma-ray computed tomography (CT) technique for the first time to investigate the hydrodynamics of spouted beds. In particular, we study the effect of particle density, particle size, bed size, and superficial gas velocity on the gas-solid cross-sectional distributions of spouted beds. The color distributions of the cross-sectional images clearly identified the three regions of spouted beds: the spout, the annulus, and the fountain regions. Interesting results and findings are presented, discussed and analyzed in the article. For example, the results demonstrated that the summation that operating spouted beds at stable spouting state would lead to achieving uniform coating layers of the particles in the TRISO fuel coating process is not adequate. Further, it has been found that increasing the superficial gas velocity much higher above the minimum spouting velocity increases the gas holdup in the annulus above the gas holdup value at the loss-packed bed state, contrary to common assumptions presented in the literature. This study represents an original experimental investigation required both for advancing the understanding of TRISO particles spouted bed and providing benchmark data to validate computational fluid dynamics (CFD).

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1. Introduction

The very high-temperature reactors (VHTRs) are the most promising candidates among the fourth generation of nuclear reactors for meeting the energy demands of the 21st century. Passive safety is one of the most important features characterizing these reactors [1]. Two types of VHTRs have been designed to be fueled by TRISO (tri-structural isotropic) fuel particles: the prismatic bed reactor and the pebble bed reactor. However, the deployment of the TRISO fuel in these reactors differs. The TRISO particles are either inserted into large hexagonal graphite fuel elements (block design) or inserted into small graphite pebbles. It has been reported that the performance and safety of these reactors significantly depend on the quality of the TRISO-coated fuel particles [2-8]. The coating layers of the TRISO particles must be of high standard quality with a very low coating failure rate [7]. Current fuel standards require that there should be no more than 1 in 10^5 coating failures on individual particles [8]. The technique used for coating the TRISO fuel particles is gas-solid spouted bed via chemical vapor deposition (CVD) method. It has been reported that the spouted bed coater is the key part of the coating process, the quality of the nuclear fuel particles produced is strongly impacted by the hydrodynamics of the gas-solid spouted beds [8, 9]. Accordingly, a fundamental understanding of the underlying phenomena of the spouted bed's hydrodynamics is essential.

Spouted beds are gas-solid contactors widely used for different applications of chemical engineering and mining processes such as gasification, polymerization, granulation, coating, and drying [10-13]. The spouted bed was originally developed in 1954 at the National Research Council of Canada by Mathur and Gishler [10, 14] to overcome some of the limitations in fluidized beds in handling heavy, coarse, and unevenly-sized particles. Key features of spouted beds are the capability to provide uniform thermal distribution, good particles mixing, and efficient contact between the solid and fluid phases. The spouted bed consists of a vertical inlet that directs gas upwards through the base center of a conical, cylindrical, or cylindrical-conical reactor, which contains a bed of particles. If the velocity of gas introduced into the bed of particles is sufficiently high, the gas carries the particles up rapidly through the bed axis

and, thus, forms a hollowed region in the center of the bed. The particles then somewhat above the bed surface, fall freely back due to gravity to the bottom of the bed in continuous circulation. During this process, a systematic cyclic movement of the solids is established and three distinct regions are thus created in spouted beds each with specific flow behavior; a spout in the center of the column where the particles are carried up by the gas to the top of the bed surface, an annulus surrounding the spout where the particles fall back to the bottom of the bed, and a fountain on the top of the spout-annulus regions in which the particles transition from the spout to the annulus. [Figure 1](#) shows a simple schematic diagram of a spouted bed (inspired from the work of Olazar et al [\[12\]](#)) and illustrates the three regions: the spout, the annulus, and the fountain. It is clear that such a flow pattern in spouted beds leads to complex recirculation, which engenders strong gas–particle interactions, allowing for many applications for this reactor.

In the coating process of the TRISO particles, the fuel kernel (UO_2) of low-enriched uranium (LEU) is coated consecutively with four layers: a porous buffer pyrolysis carbon layer (buffer PyC), an inner dense pyrolysis carbon layer (IPyC), a silicon carbide layer (SiC), and an outer dense pyrolysis carbon layer (OPyC). As mentioned earlier, all these layers are produced in gas-solid spouted beds via the chemical vapor deposition process. However, the sizes and densities of the kernels, and the coating layers, are of prime interest with respect to the hydrodynamics of spouted beds during the coating process. The density of the kernel UO_2 ($\rho_p \sim 10.80 \text{ g/cm}^3$) is noticeably bigger than those of general particles found in chemical engineering study, for example glass beads ($\rho_p \sim 2.60 \text{ g/cm}^3$). Therefore, the impact of particle density on spouting behavior is essential and should be investigated systematically [\[15\]](#). Since the layers compact together to form a non-homogenous mixture of multiple materials, the size and density of the particles during the coating process are not constant and vary with each layer deposition, as shown in [Figure 2](#) [\[9\]](#). The density of the kernel is approximately 10.8 g/cm^3 and the size is 0.5 mm . The average density of the particle is then reduced substantially to 4.1 g/cm^3 when the particle is coated by just the first layer (low-density pyrolytic carbon), and the size increase to 0.68 mm . This process takes place for all the coating layers resulting in a final particle density of about 2.65 g/cm^3 and a size of about 1 mm . It is clear that the change

of particle properties will undoubtedly have a significant effect on the hydrodynamics of the spouted bed and the coating performance. The issue becomes more complex when the probability of successfully coating a layer depends on the success of the preceding coating. Moreover, the outlooks for future fuel-coating technologies and applications are further complicated by the fact that many new concepts will involve fuel kernels of various sizes, compositions, and densities. Having a fundamental understanding of the gas and solids dynamics is critical for evaluating gas-solid contacting efficiency in spouted beds [16].

In the literature, only few studies on spouted beds operating with heavy particles of density above 5 g/cm^3 , mostly encountered in the TRISO coating process were, so far, reported [17, 18]. Zhou and Bruns [17] experimentally studied the minimum spouting gas velocity of zirconia particles (ZrO_2 , $\rho_p = 5.89 \text{ g/cm}^3$) in shallow spouted beds, with the help of pressure drop sensors. They established new correlations for estimating the minimum spouting velocity based on experimental data. In addition, Sari et al. [18] investigated the minimum spouting gas velocity of ZrO_2 particles ($\rho_p = 6.05 \text{ g/cm}^3$, $d_p = 0.5, 1 \text{ mm}$) by means of pressure drop measurements in spouted beds with a diameter of 150 mm and cone angles of 30° , 45° , and 60° . The investigation indicated that the steady spouting state exists in the range of $1.2U_{ms}$ – $2.1U_{ms}$, beyond which jet spouting begins; U_{ms} is the minimum spouting velocity. The above studies only focused on the minimum spouting velocity and one type of high-density particles. To date, no comprehensive studies about the impacts of high-density particles on local hydrodynamic parameters of spouted beds have been reported. Pannala et al. [8] used the Multiphase Flow with Interphase eXchanges (MFIx) computer code to simulate the spouting process of zirconia particles in 2-dimensional rectangular and conical-bottomed spouted beds. This model treats phases as continuous and interpenetrating, yielding a granular layer pseudo-fluid, which requires the definition of some properties such as viscosity bulk, shear the granular phase and pressure granular stage properties present in the momentum balance equation. Absence or unavailability of advanced measurement techniques for experimental validation were the reasons for why they simulated 2-dimensional spouted beds instead of fully 3-dimensional spouted beds. They pointed out that experimental

studies are essential for better optimization and validation of more accurate computational modeling of spouted beds. Furthermore, Liu et al. [9] used pressure sensors at both ends of the spouted beds to establish a relationship between the pressure drop change prior and following the coating process of each layer in the real TRISO fuel-coating process. The authors found that the pressure drop change occurred simultaneously with the change of particle properties during the successive coating of each layer. They also pointed out that experimental investigations are needed to quantify specific influences of key parameters on local spouted bed hydrodynamics. Liu et al. [15] used the CFD–DEM (Computational Fluid Dynamics–Discrete Element Model) coupling simulation method to examine the fluidization behavior of particles with different densities in the spouted bed. The simulation results revealed that the spouting behavior of the particles is incoherent and periodic when the spouting state is stable. They reported that the stable spouting range expands with particle density increases.

Taking into account the complexity of the two-phase flow through a spouted bed, a comprehensive knowledge of the hydrodynamics involved has not been achieved to date. Many experimental and computational works on the hydrodynamics and flow field of gas-solid spouted beds, in general, have been reported [19-38]. The reported studies used various techniques, such as direct visualization, pressure measurements and various probes to measure global dynamic parameters (e.g., spout diameter, spout shape, spoutable height, fountain height, etc.). Furthermore, some of them employed intrusive techniques such as optical and capacitance probes for local point measurement. Both optical fiber and static pressure probes have been widely used for hydrodynamic characterization of the spouted bed systems. However, these techniques have several limitations, a major one being the local disturbance of the gas-solid flow caused by the probe [39]. Moreover, the conditions in all of these studies focus on low density particles ($\rho_p < 3.0 \text{ g/cm}^3$). It is then necessary to acquire accurate knowledge on the crucial parameters that characterize the hydrodynamics behavior of spouted beds, such as cross-sectional phases' distributions, holdups radial profiles along the bed height, 3-D solid velocity fields, turbulent parameters, gas dispersion, mixing behavior, and others. Recently, it has become possible to assess these key parameters and to improve well-

developed models and correlations for spouted beds using advanced non-invasive velocimetry or tomography techniques. In our laboratory, the mentioned assessment has been achieved using advanced non-invasive techniques: Gamma ray computed tomography (CT) and Radioactive particles tracking (RPT) techniques. With these techniques, we were able to address important issues related to the scale up of spouted beds [40, 41]. It has been proven through practical experimentation and research studies, that CT and RPT are the most promising technologies for visualizing multi-phase flow systems (i.e. gas-liquid, gas-solid, gas-liquid-solid phase) [42, 43].

In the present contribution, we aim to provide an experimental investigation of the hydrodynamics of gas-solid spouted beds at cold conditions using advance non-invasive technique; gamma ray computed tomography (CT). The gas-solid distribution and its variations have a strong impact on the performance of the spouted beds. Assessment of phase (gas or solids) distributions in spouted beds is of primary concern for accurate design and control of the TRISO fuel-coating process. For example, knowledge of the gas and solids holdup distributions would define parameters such as the spouting conditions, the coating gas composition, and the coating temperature, which will, in turn, result in a higher probability of coating success. The coating layers deposited on the particles depend on how and where the particles are present in the bed. In spouted beds, the solids holdup distributions (or gas holdup) differ from region to region and these differences cause changes in gas-solid interactions and, above all, changes in the value of the absorption and the conversion of gaseous reactants. In this study, the effects of the particle properties and operating parameters on the gas-solid distribution of spouted beds are the focus. It is hoped that the originality of the results and the analysis of them presented here will provide the basis for assessing design and control processes of TRISO fuel coaters, and contribute to other spouted bed applications. This study will provide reliable and valuable benchmark data for the assessment and validation of CFD models and their closures. Improved validation of CFD models for reliable and safe design, scale-up, and process optimization will ensure proper performance and operation of TRISO fuel coaters. Finally, successful implementation of the CT as a non-invasive

technique on the cold-flow coater will lay the ground for its implementation in hot pilot plants and industrial units.

2. Experimental set-up and measurement technique

2.1. Experimental set-up

The actual experimental works, similar to that used by Ali et al. [40, 41], consist of two different size of fully cylindrical-conical base spouted beds with inner diameter of 0.076 m and 0.152 m. The deployed spouted beds were fabricated from Plexiglas material and each unit consisted of one-piece column attached to a conical base. The schematic diagrams of the two spouted beds employed are shown in Figure 3. Both columns were specified and designed to be geometrically similar with 1.14 m height and conical base of 60-degree angle. The two columns were constructed without having any ports or connections on the wall surface to avoid the possibility of any non-symmetric issues for the columns, which in turn mess and complicate the CT reconstructions procedure. At the bottom of the conical base, a gas distributor made of stainless steel was placed to pass through the gas flow inside the column. The diameters of the inlet orifice are 9.5 mm and 19.1 mm for the 0.072 m and 0.152 m spouted beds respectively. The gas used was dry compressed air supplied from an industrial compressor with capacity up to 735 CFM and 200 Psig. The air was filtered before being passed through into the column at ambient temperature ($T= 20^{\circ}\text{C}$). After the flow pass through the piping components of the compressor, the rate of the flow is regulated, using a pressure regulator and rotameter setup made by parallel connecting of two rotameters. The gas flow rates were maintained at the desired operating condition with the aid of the rotameters and a set of gauge valves and were introduced into the spouted beds through the inlet orifice (gas distributor). All the experiments were carried out at the ambient temperature and the atmospheric pressure. The design of the operating conditions for spouting stability was described in detail in previous paper [41], and was based on the findings of [10, 37, 44, 45].

Four different particulate materials, nearly spherical in shape with narrow size distribution, were employed in the experiments: steel particles of mean diameter 1 mm and 2 mm and glass beads of mean diameter 1 mm and 2 mm. The advantage of using steel particles is their high density allows the determination of the hydrodynamics of spouted beds of heavy particles and the way they differ from relatively spouted beds of light particles. The glass particles were chosen to enable the comparison of the results of this study with those reported in the literature where glass beds are largely used for fluidization studies. Another reason why we selected the steel and glass particles is the density ratio between the two materials ($\rho_{\text{steel}} / \rho_{\text{glass}} \sim 2.96$). This ratio made the two materials excellent choices, since in the actual spouted bed nuclear fuel coaters a close ratio is obtained after the coating of the first layer ($\rho_{\text{kernel-Uo2}} / \rho_{\text{coated-kernel}} \sim 2.63$). Therefore, the results of the two materials will be valuable for designing and understanding, and can be linked to the spouted bed nuclear fuel coaters. The relevant properties of the particulate materials are listed in [Table 1](#). The stagnant bed heights range from 0.14 m to 0.32 m. The superficial gas velocities vary from $1.1U_{\text{ms}}$ to $1.4U_{\text{ms}}$, where U_{ms} is the minimum superficial gas velocity required to obtain the onset external spouting state in spouted beds. Below this velocity, solids circulation is absent, and, consequently, all the three regions in spouted beds are not fully created. The minimum spouting velocity can be obtained experimentally or theoretically, and it is a function of the physical properties of the particles used (density, size, shape), bed geometry (column diameter, inlet diameter, cone angle), static bed height, and gas properties (density, viscosity). The minimum spouting velocities were measured experimentally and compared with the well-known correlation available in the literature by Mathur and Gishler for deep spouted beds ($H_o/D_c > 1$) (Eq.(1)) [[14](#)]. The measured values are found close to the estimayed ones by the correlation with average percentage increase of 7.31%.

$$U_{ms} = \left(\frac{d_p}{D_c} \right) \left(\frac{D_i}{D_c} \right)^{\frac{1}{3}} \sqrt{\frac{2gH_o(\rho_p - \rho_g)}{\rho_g}} \quad (1)$$

2.2. Dual source gamma-ray computed tomography (DSCT).

Dual Source Gamma-ray Computed Tomography (DSCT) is a systemized technique used to obtain measurements of cross-sectional phase distribution of two-phase or three-phase flow systems (gas-liquid, gas-solid, and gas-liquid-solids). The CT has been employed for visualization of different multiphase flow systems [46-60]. The details of the hardware and software information of the DSCT have already been elaborated in detail by Varma et al. [50]. The DSCT setup consists of a selection of collimated detectors and opposing sources present on a platform that can rotate 360° around the column that needs to be scanned. It provides 197 views in each scan and 21 projections in each view. Figure 4 shows a typical 3-dimensional (3D) model of the DSCT technique's platform. This platform can move up and down to capture the vertical view of the column to generate 2-dimensional (2D) and 3-dimensional (3D) images. The DSCT setup consists of two sealed gamma ray sources. Each source is housed on a collimated device in such a way to provide a fan-beam shape with a thickness of about 3 mm and a fan angle of 40° .

The radioactive isotopes used in the DSCT are Cs-137 and Co-60 with initial activity of about, 200 and 300 mCi (millicurie), respectively. The Cesium (Cs-137) and Cobalt (Co-60) were the most frequently used isotopes as gamma-ray sources. The Cs-137 is commonly used in nuclear gauging applications in industries because it has a long half-life, it does not scatter interference of photons of other energies and emits single clear photo peak. Co-60 is also widely used because of its small scatter interference of signals. Based on the number of sources used in the DSCT system, it can be called as a single source in a case it is one or a dual source in a case the two sources are used. In this work, only Cs-137 was used to distinguish between the solid and gas phases of spouted beds and the technique called CT. Opposite to each source, an array of 15 sodium iodide (NaI) scintillation detectors is located on an arch plate to collect the unattenuated gamma rays

passing through the column. The detectors are collimated by a 2 mm slit width lead collimator, attached on the crystal of each detector. The CT has a connection to a power supply that provides the charge to excite the detectors. The detectors absorb the fallen radiation and release it as light photons that hit photo-sensitive cathodes and multiplier tubes. The free electrons that arise in the process are multiplied and sent to the output as current. The signals from the detectors are amplified by a timing amplifier to increase the amplitude from the detectors. The NaI detectors used were Canberra Model 802 scintillation detectors. Each detector in the system consist of NaI (TI) scintillation crystal (2×2 inches), a photomultiplier tube (PMT), a light shield, aluminum housing, and a 14 connections pins. The High voltage (HV) applied for the PMT of each detector was 900V. The detected signals coming from the detectors were discriminated from background radiation and recorded by a fast-timing multilevel discriminator and a scaler respectively.

The procedure for obtaining the cross-sectional values of gas and solids consists of multiple steps. Some of these steps include: the CT scans, averaging of raw data, assignment of initial guess values, and the reconstruction process. Complete sets of CT scans were carried out in each experiment to obtain the phase density distribution of gas-solid spouted beds. These scans include those when the column is under the desired conditions of the gas-solids spouted bed operation (i.e. The actual test), the column is filled with solids as a packed bed (i.e. Spouting gas is absent), and the reference one taken when the column is empty (i.e. Full of air only). The counts for each projection were collected by the collimated detectors for 60 samples at 20 Hz to improve accuracy. The means for the data points of each projection, for each projection method, and all scans performed are obtained. The mean value for all the counts based on multiple samples of a given projection is used to process the data. The reconstruction process was conducted using the Alternating Minimization method [50, 61]. The procedures for the reconstruction process used to obtain gas and solids holdups distributions in the present spouted beds have already been elaborated in recent work [41]. The reconstruction domain of the beds was discretized by pixels. Cross-section of a given spouted bed column was encompassed by a square matrix of pixels with dimensions of (n) rows $\times$ (m)

columns pixels. The corresponding size of pixels was 80 x 80 for the 0.152 m and 0.076 m spouted beds. Hence, each pixel represents approximately 2 mm in size in case of the 0.152 m spouted bed, and 1 mm in size in case of the 0.076 m spouted bed. The accuracy of the CT technique was proven by implementing test scans for standard specimen, so called “phantom”. The phantom is made of two concentric Plexiglas cylinders with outer diameter of 0.152 m, and inner diameter of 0.076 m where one of them was filled with water. The reconstructed phantom have shown the measured linear attenuation of water and the dimensions of the cylinders in comparison to the actual values were within up to 1%. In order to ensure the reproducibility of the CT technique and for accurate comparability between the investigated conditions, a repeated set of scans was carried out at selected level, using similar bed operating conditions, and the method was found to give excellent reproducible results. For the sake of brevity, the reproducible results of the CT technique are not shown here but the reader can refer to previous paper [\[41\]](#).

3. Results and discussion

The cross-sectional distribution of the phases' holdups of gas-solid spouted beds was studied by CT. The aim is the establishment of a map of local bed holdups in the three regions of spouted beds, which allow for assessing the design of the TRISO fuel coating process. More precisely, we investigated the solids holdup distributions for spouted beds of particles of different densities and sizes and beds of different sizes. The results highlight key phenomena governing the gas-solids distributions, and prove the significance of the impact of the gas dynamics on the hydrodynamics of spouted beds. The findings will lead to further discussion about the influence of the U/U_{ms} on the gas and solids holdups distributions of spouted beds. The discussion on the results will involve two perspectives points: the spouted beds hydrodynamics in general and the spouted beds hydrodynamics from the TRISO fuel coating viewpoint.

3.1. Effects of particle density and size

Figure 5 and Figure 6 present the measured time averaged cross-sectional distributions of the solids holdups as a function of bed material using the CT. The solids used are high-density steel particles ($\rho_p = 7.4 \text{ g/cm}^3$) and glass beads ($\rho_p = 2.5 \text{ g/cm}^3$). The scale-bar on the right of the images indicates the fraction of the solids holdup. Red color is an indicator of a higher solids holdup, whereas the blue is an indicator of a lower solids holdup. The color scale used in the figures is kept constant for convenience of comparison between the images. The color distributions of the images clearly highlight the three regions of spouted beds: the spout, the annulus, and the fountain regions. Overall, the cross-sectional distribution images provide a quantitative picture of the way the solids are distributed in spouted beds. It should be pointed out that the cross-sectional phase distribution reported in this study is the only quantitative study in the literature on

the gas-solid spouted beds. Exception is made for recent work of our group [41]. The experimental results in Figures 6 and 5 are qualitatively similar to those corresponding to other examined systems here and exhibit the significant variations of the solids holdup with bed position. The solids holdup distributions of the steel and the glass vary in the same way with respect to the z/D ratio. For each material, the solids holdup increases with increasing the bed height and the reverse is true for the gas holdup (gas holdup = 1 - solids holdup). This increase occurs when some solids enter the spout from the annulus at levels below the bed surface. The variation of the solids holdup with the radial position is pronounced. At each z/D level, solids holdup are seen to be low near the axis of the spout and increase, very slightly near the axis, then very sharply at the spout-annulus interface. In the annulus, the solids holdup distribution is almost unchanged along the bed height. At the column wall, the solids holdup is seen to be unchanged. It can be observed in Figure 5 ($D_c = 0.076$ m, $\gamma = 60^\circ$, $D_i = 9.5$ mm, $d_p = 1$ mm; $H_o = 160$ mm, $U/U_{ms} = 1.1$), based on the color scale located at the right of the images that as material density increases, solids holdup decreases. Steel particles demonstrate lower solids holdup in the spout than the glass particles at the same U/U_{ms} ratio. This solids holdup decrease is due to the increase in the minimum spouting velocity of the solids, when the value of material density increases. It means that at the same U/U_{ms} ratio between the two materials, the glass particles are more existing at a lower superficial gas velocity. The same finding is observed for the 0.152 m spouted bed column in Figure 6 ($D_c = 0.152$ m, $\gamma = 60^\circ$, $D_i = 19.1$ mm, $d_p = 2$ mm; $H_o = 160$ mm, $U/U_{ms} = 1.1$).

In order to verify the qualitative observations made between the different bed material densities, quantitative comparisons of solids holdup between materials are also presented in this section. From Figure 5 and Figure 6, it can be observed that the cross-sectional solids holdup distributions in spouted beds are axi-symmetrical. Therefore, one is justified representing the cross-sectional distribution through azimuthal averaging of the two-dimensional data. The cross-sectional images in Figure 5 and Figure 6 are azimuthally averaged to obtain the radial profiles in Figure 7 and Figure 8, respectively. Figure 7 shows the comparison of the solids holdup radial profiles of different material studied under selected spouted bed system ($D_c = 0.076$ m, $\gamma = 60^\circ$, $D_i = 9.5$ mm, $d_p = 1$ mm;

$H_0 = 160$ mm, $U/U_{ms} = 1.1$) at different z/D levels. In the spout, which is characterized as a gas-solid riser the solids holdup is lower near the gas inlet and increase as the height increases along the bed. As pointed out earlier, this increase in solids holdup is due to the entrainment of particles from the annulus along the height of the bed, in agreement with the observations of [20, 22, 25, 62]. [10, 63] stated that the cross-sectional average gas holdup decrease with increasing bed height, and the reverse is true for the solids holdup. Indeed, Roy et al. [64] examined a bimodal cycle time distribution of the radioactive tracer. They showed that there are two main solid exchange regions. One at the conical base and the other just below the bed surface. Some authors reported that there is a loss of gas to the annulus at level below the bed surface resulting the increase on the solids holdup [62]. This loss of gas is not noticed for the investigated conditions shown in Figure 7 and Figure 8. The variation on the solids holdup profiles is not pronounced along the bed height of the annulus. The solids holdup profiles in the annulus are almost unchanged along the bed height, with an average value of 0.59. Hence, particle-particle contacts and interaction forces cannot be ignored in the annulus. It is also to be noticed that the shape of the solids holdup profiles in the spout region is much closer to a parabolic shape. The profiles show that the solids holdup is low near the axis of the bed and increase radially toward the spout-annulus interface. The experimental results in Figure 7 and Figure 8 present the great variation of solids holdup in the spout with bed position, either the longitudinal or the radial directions. The profiles obtained here are in excellent agreement with the recent works [41, 65, 66]. Comparison to be made using these results related to correlations prediction of solids holdup are restricted. To the authors' knowledge, it seems that there are no correlations predicting solids holdup in spouted beds available in the literature. The only correlations available in the literature are to predict the gas holdup for conical spouted beds [22, 25]. However, these correlations of conical beds (made with fully conical geometry) cannot be applied to the conventional spouted beds, also known as conical-cylindrical spouted bed, due to geometrical differences between the two beds [67-69]. In the fountain, the solids holdup profiles differ from those collected in the other regions of spouted bed. The reason is the unique motion of the particles in this region, which can be described as a bridge to redirect the particles from the spout back into the annulus. The solids holdup in the

fountain is crucial for the solids distribution from the fountain into the annulus, and the solids circulation in the entire bed. The profiles show the solids holdup is higher near the core of the fountain and decrease radially toward the outer region of the fountain. It has been reported the importance of the fountain in promote the particle segregation in spouted beds [70, 71]. Waldie et al. [72] also considered the significance of the fountain when the spouted bed is used as a coater, the interaction of a feed spray with particles in the fountain is impacted by the gas-solids distribution in the fountain. Hence, attention should be paid in future for the fountain in the coating process of TRISO fuel particles using spouted beds.

As shown in Figure 7, as the density decreases the solid holdup increases, confirming what is found in the cross-sectional images. The effect of increasing the solids density is a decrease in the solids holdup. Glass beads have higher solids holdup in the spout than steel particles when the two systems are applied at the same U/U_{ms} ratio. The reason is mainly the increase of the minimum spouting velocity of steel particles. This finding about the spout is in agreement with the experimental results of San José et al. [25], who used optical probe in conical spouted beds. San José et al. [25] reported that the gas holdup increases when the minimum spouting velocity increases and when the value of the particle density increases. However, the reverse is true for the solids holdup. In Figure 7, the average percentage difference between the profiles of the two materials in the spout is 54.62%, 46.96%, 37.39%, respectively, at z/D levels of 0.8, 0.9, and 1.1. The differences in the solids holdup of the two materials can be explained as follows. Since the superficial gas velocity applied for the steel particles is higher than that of glass, the particles with the smaller density will less drag in the spout than the particles of the greater density. As a consequence, they will lag behind the air flow in the spout region and in the core of the fountain as well. Thus, the particles with the smaller density are expected to have the lower axial velocity and the longer time to travel through the spout than the particles with the greater density. Accordingly, the particles with the smaller density are more presented into the spout region and solids holdup increases. The results also can give the thought that the glass particles fed more into the spout region than the steel particles. However, no noticeable differences were observed for the solids holdup

profiles of the two materials in the annulus region. This is primarily attributed to the very slow movements of the particles in the annulus, where the particles move slowly downward in this region as in moving packed bed. The average percentage difference between the profiles in the annulus is small. The results indicate that the solids holdup distributions in the annulus region are less dependent on the material density. In the fountain, the glass particles demonstrated higher solids holdup than steel particles. The average percentage difference between the profiles of the two materials in the fountain is 34.44%. The results presented in [Figure 8](#) demonstrate the same findings as in [Figure 7](#), despite the differences in the bed size and the spouting conditions. In [Figure 8](#), the glass particles show higher solids holdup values than the steel particles, both in the spout or the fountain. The average percentage difference between the profiles of the two materials in the spout is 25.09%, 28.9%, 21.9%, respectively, at z/D levels of 0.8, 0.9, and 1.1. The radial profiles of the two materials compared in the annulus region have small or no deviations. The average percentage difference between the profiles of the two materials in the fountain is 18.99%.

The particle size has an essential role in gas-solids spouted beds and, therefore, in the coating process of TRISO particles. For example, the heat transfer from the wall-to-the bed, from solid-to-solid surface, and from the gas phase-to-the solids in spouted beds largely depend on the distribution of the solids holdup and increase with increasing the particle size. In addition, the amount of the coating layers captured by the particles and quality of that layers are also dependent on the size of the particles (i.e. Surface area of the particles). The size of the particles varies during the coating process of the TRISO particles. Thus, additional information on the solids holdup distributions were obtained by examining the effect of the particle size. [Figure 9](#) and [Figure 10](#) show the comparisons of the solids holdup radial profiles of material of different sizes and same density under selected spouted bed systems. The results concerning the impact of the particle size in the annulus are found less accentuated and not shown in the figures. The influence of the particle size on the solids holdup is quite evident in the spout region. In [Figure 9](#), the average percentage difference between the profiles of the different sizes particles in the spout is 9.74%, 5.85%, 7.37%, respectively, at z/D levels of 0.8, 0.9, and 1.1. The results

suggest that increasing the particles size leads to a decrease in the solids holdup in the spout at the same U/U_{ms} . This decrease is caused by the increase of the minimum spouting velocity when the particle size increases. The particles with the smaller size are expected to be present more in the spout and, therefore, have a longer contact with the gas phase. In [Figure 10](#), the average percentage difference between the profiles of particles of different sizes in the spout is 9.53%, 7.54%, 6.41%, respectively, at z/D levels of 0.8, 0.9, and 1.1. Comparing the deviations here with those for the different densities, we found that the particle size has less impact on the solids holdup distributions than the particle density for the range investigated.

The results discussed above demonstrate that the changes in the properties of the particles must be accompanied by appropriate changes in the input process of the spouting conditions to produce particles with uniform coating layers. The assumption that stable spouting mode would result in uniform coating layers for the particles is not adequate. The results show that even though the particles were operated at stable spouting state and same U/U_{ms} , differences were obtained in the solids holdup distributions in the spout and the fountain regions. The coating formation of particles in a spouted bed is ultimately a function, but not limited to the solids holdup distributions of the particles. Recently, Liu et al. [\[15\]](#) used the CFD–DEM coupling simulation method to study the fluidization behavior of particle with different densities in the spouted bed. Liu et al. [\[15\]](#) reported that the stable spouting range expands when the particle density increases. Hence, there is a larger velocity adjust range to be selected from initial spouting to the end of stable spouting state. Furthermore, the authors determined the power spectral density distribution of the time domain signal of the axial instantaneous particle velocity under different U/U_{ms} ratio for various material densities. They reported that the dominant frequency of the particle spouting process and the particle residence time distribution of various densities will be almost the same in the spout region, even if the particle density increases, as long as U/U_{ms} is not changed. In addition, they stated that with the consistent residence time of all the particles in the spout region, the layers will uniformly deposit on the surface of all particles. The results reported in our work do not comply with the findings of Liu et al. [\[15\]](#). As shown in the figures, when two materials

are operated at the same U/U_{ms} , solid holdup distributions of the two materials are different. Therefore, the mass of materials deposited on the particles will be varied due to the variation on the number of particles present in the spout and in the fountain at a given time. Recently, the preference has been to move towards coating the TRISO layers continuously, without unloading the particles after each coating step to avoid smooth and sharp interface between the layers and to avoid mechanical shocks caused by unloading procedure of the particles [73]. Therefore, it is necessary to optimize the spouting conditions of the particles in the intermediate coating process of each layer due to the change of the properties of the particles, in agreement with the findings of Minato et al. [73]. The results reported here also demonstrate that the solids flow is strongly influenced by the forces exerted by the gas on the solid particles. Indeed, Al-Dahhan et al. [65, 66, 74] considered the significance of the gas dynamics in gas-solids spouted beds. [65, 66, 74] stated that the gas dynamic dictates the hydrodynamic of gas-solid spouted beds, and they used this assumption to develop a new mechanistic scale-up approach based on matching gas holdup profiles between two different sizes of beds. This is also evident in the results reported here, since the deviations between the profiles of the two materials are prominent in the spout region, as the spout region is predominantly dominated by the gas phase.

3.2. Effect of bed size

The hydrodynamics behavior of spouted beds differs with scale. Hence, it is not always possible to determine the hydrodynamics of large scale spouted beds, depending on the knowledge of smaller ones. More precisely, it means that if bed diameter is changed most of the other parameters will change concordantly. In the literature, only a few works have addressed the scale up spouted beds [40, 41, 66, 75, 76]. The last works have given a qualitative and quantitative studies on the scale-up of spouted beds. These studies make it apparent that the effect of the bed diameter needs further investigation. Therefore, it is important to obtain further fundamental insights in the effect of the bed diameter to formulate precise hypotheses on their behavior of spouted beds reactors. Figure 11 and Figure 12 show the measured time averaged cross-sectional distributions of the solids holdup as a function of bed size. The bed sizes used are 0.076 m and 0.152 m ID (inner diameter) spouted beds. The first observation inferred from the images is that the spouting structure of both sizes of the beds is qualitatively similar, with the agreement with the results of [41, 66] for scale-up of spouted beds at different conditions. However, based on the color distributions of the images, the solids holdup increases as the bed size increases. The 0.152 m ID spouted bed has higher solids holdup than the 0.076 m ID

spouted beds when the two beds are applied at the same U/U_{ms} and H_o . The solids holdup is higher in the 0.152 m ID spouted bed because the U_{ms} value for the 0.152 m ID spouted bed is lower than that for the 0.076 m ID spouted bed. [41] reported higher solids holdup for bed of large size at different operating conditions. Figure 13 and Figure 14 show the comparisons of the solids holdup radial profiles for beds of different sizes under selected spouted bed conditions. The comparison of solid holdup profiles in 0.076 m and 0.152 m spouted beds demonstrates that the solids holdup in 0.152 m spouted bed is higher than that in 0.076 m spouted bed. From the result, the only deviation of the solids holdup profiles between the two beds was observed in the spout. However, in the annulus region, unchanged profiles are achieved for both beds. The movement of the particles in the annulus of both beds is the same, the particles move slowly and downward as in moving packed bed. In Figure 13, the average percentage difference between the profiles of the different beds sizes in the spout is 24.34%, 16.17%, and 23.91%, respectively, at z/H_o levels of 0.71, 0.85, and 0.96. The average percentage difference between the profiles in the annulus is small. In Figure 14, the average percentage difference between the profiles of the different beds sizes in the spout is 37.68%, 19.12%, and 15.06%, respectively, at z/H_o levels of 0.71, 0.85, and 0.96.

3.3. Effect of superficial gas velocity

In the literature, it is commonly assumed that the gas holdup in the annulus region of spouted beds is constant along the bed height and equal to the gas holdup value at loose-packed bed state ($\epsilon_o = 0.411$), where there are particles but the gas flows through the voids between the particles without disturbing them significantly [14, 77-80]. Accordingly, different measurements techniques, CFD models, and closures for simulating spouted beds, including studies targeting TRISO fuel coating, were based on this assumption. However, the question whether there are variations in the gas holdup in the annulus or not is not completely covered. Hence, further investigation was conducted to resolve this question, along with the influence of the U/U_{ms} on the spout and annulus regions, taking into consideration the advantage of the non-invasive measurements of the CT technique without disturbing the flows in spouted beds. Figure 15 show the time-averaged cross-sectional distribution and the corresponding frequency distribution for gas holdup of selected spouted bed system ($D_c = 0.152$ m, $\gamma = 60^\circ$, $D_i = 19.5$ mm, solids: glass beads, $d_p = 2$ mm; $H_o = 325$ mm) at z/D level of 1.1 and different U/U_{ms} ratios. At no time during the experiments, the annulus appeared to be fluidized. The images show the effect of the U/U_{ms} ratio on the gas holdup distributions, as the gas holdup at many

local positions in the spout and the annulus (in strict sense) regions increased with increasing the U/U_{ms} ratio. A more detailed view of the gas holdup measurements can be obtained from the statistical analysis. Pixels outside the spouted bed diameter (i.e. pixels with gas holdup equal to one) were not considered in the normalized frequency distribution for a case of comparison. The mean and the mode of the distribution are only affected by the magnitude of the gas holdup values in the spout and the annulus regions. The normalized frequency distribution is predominant at the gas holdup value in the annulus and skews to the right where the frequency distributions are small in the spout. The skewness distribution (non-symmetric) is expected for spouted beds due to lower and upper gas holdup values on the data. Also, the skewness distribution of the peaks further confirms the divergence between the spout and the annulus regions in spouted beds. The mean values of the frequency distribution below the images disclose that the gas holdup increases in the entire bed. As the U/U_{ms} ratio increases, the mean value of the gas holdup cross-sectional distributions increases. At $U/U_{ms} = 1.1$, the mean of gas holdup values is 0.429, above to the mean gas holdup value at the loose-packed bed state ($\epsilon_0 = 0.411$). However, this value increased sustainably from 0.429 to 0.473 when the U/U_{ms} ratio increased from 1.1 to 1.4. Increasing the U/U_{ms} ratio changes the frequency distribution to higher values and shifts the peak plateau to the right past the gas holdup value at the loose-packed bed state. This phenomena is more obvious when one compare the peak plateau at $U/U_{ms} = 1.1$ and $U/U_{ms} = 1.4$. In addition, the standard deviation increases monotonically with increasing the U/U_{ms} ratio. Nevertheless, one can argue that the gas holdup cross-sectional distributions in the annulus remained non-fluidized for the investigated U/U_{ms} ratios. Therefore, diverting the flow of air to the annulus should only have contributed to increase the gas holdup, with little effect on the structure of the annulus.

For further comparison of the results, the results of solids holdup cross-sectional distributions are also presented here. The effect of the U/U_{ms} ratio on the cross sectional distribution of the solids holdup at z/D level of 1.1 is shown in [Figure 16](#). The images demonstrate that the solids holdup distribution is nearly symmetrical around the bed axis. Such symmetry exists at $U/U_{ms} = 1.1$ and even at the higher U/U_{ms} ratio. Nonetheless, it is

evident from the images that increasing the U/U_{ms} shifts the solids holdup to decrease whether in the spout or in the annulus. The images also show the spout size increase with increasing the U/U_{ms} , in qualitative agreement with [19, 81, 82]. The effect of increasing the U/U_{ms} ratio in spouted beds can be explained by observing it as a decrease in the solids holdup due to the increase in the gas holdup whether in the spout or in the annulus when the superficial gas velocity is increased. In the annulus, the voids between the particles at many local positions increase with increasing the U/U_{ms} ratio, confirming what is presented in Figure 15. This finding is more pronounced at $U/U_{ms} = 1.4$, where pixels with yellow colors increased in the annulus region. The mean of the solids holdup cross-sectional distribution is 0.571, 0.565, 0.547, and 0.526, respectively at U/U_{ms} ratio equals to 1.1, 1.2, 1.3, and 1.4. The mean of the solids holdup distribution is affected by the magnitude of the phase holdup values in both regions (the spout and the annulus). Consequently, in order to properly analyze the results, the data of each region in spouted bed must be excluded from the other and analyzed separately. We will restrict the following discussions on the gas holdup profiles.

Figure 17 shows the radial profiles of the gas holdup in the annulus region along the bed height at different U/U_{ms} ratios for selected spouted bed system ($D_c = 0.152$ m, $\gamma = 60^\circ$, $D_i = 19.5$ mm, solids: glass beads, $d_p = 2$ mm; $H_o = 325$ mm) at different z/D levels. At $z/D = 1.1$, the average gas holdup in the annulus is 0.414 at $U/U_{ms} = 1.1$, is 0.413 at $U/U_{ms} = 1.2$, is 0.431, at $U/U_{ms} = 1.3$, and 0.441 at $U/U_{ms} = 1.4$. For only one level, ($z/D = 1.1$), the average gas holdup in the annulus increased by 4.87%, and 7.56%, respectively, for U/U_{ms} equal to 1.3, and 1.4, above loose-packed bed value. The results show that the gas holdup in the annulus increases with increasing the ratio of U/U_{ms} despite the rise in the level of the bed height; increasing the U/U_{ms} ratio allows for additional gas flowing between the particles. The results demonstrate that the conventional assumption that the gas holdup in the annulus is constant and equals to the loose-packed bed value is not completely accurate. This finding is in line with the finding reported in the literature [20] and contradicts the common spread assumption that the gas holdup in the annulus equals to the loose-packed bed value. [20] measured the gas holdup in the annulus using optical probe for conical-cylindrical spouted bed under comparable conditions ($D_c = 0.152$ m, $\gamma =$

60°, $D_i = 19.5$ mm, solids: glass beads, $d_p = 1.14$ mm; $H_o = 325$ mm, $U/U_{ms} = 1.1, 1.2,$ and 1.3) similar to the ones presented in [Figure 17](#). They reported that the gas holdup in most of the annulus is above the loose-packed bed value. In their work, the mean of the gas holdup values in the annulus increased by 2.5%, 4.9%, and 11.6% higher than the loose-packed bed value of 0.412, respectively, at $U/U_{ms} = 1.1, 1.2,$ and 1.3 . It should be noted that the increase on the gas holdup values in their work was observed starting from $U/U_{ms} = 1.1$, while in this work it was observed at $U/U_{ms} = 1.2$. The above assumption, in fact, simplifies the actual process occurring in the annulus of spouted beds. The annulus does not remain at the loose-packed bed gas holdup. Rather, it can include additional gas flowing between the particles, therefore resulting in the dynamic distribution of the two phases of spouted beds. This distribution potentially impacts reactions and transfer rates of the mass and heat in spouted beds. Hence, findings, measurements techniques, CFD models, and the reported closures for simulating spouted beds based on this assumption can be challenged due to the accuracy. The fluid dynamic in spouted beds is highly sensitive to the variations of the gas holdup. Thus, simulations of different conditions of gas holdup distribution can lead to different fluid dynamic profiles. So far, despite the attempts to apply the CFD models [[15](#), [26](#), [28-32](#), [83-87](#)] on simulating spouted beds, detailed validations of these models and closures against detailed local hydrodynamics of the annulus has not yet been covered. Accordingly, future models are required to pay attention on the annulus of spouted beds and prove that they captured the interplay phenomena and parameters. From the TRISO fuel coating point of view, the results suggest that the higher the U/U_{ms} applied on the spouted beds, the more of reactive gas will escape from the spout to the annulus or vice versa from the annulus to the spout. Thus, contact between the particles and the reactants gaseous may take place in the annulus, whereas typically it is assumed to be in the spout.

The effect of the U/U_{ms} on the gas holdup profiles in the spout for selected spouted bed system ($D_c = 152.4$ mm, $\gamma = 60^\circ$, $D_i = 19.5$ mm, solids: glass beads, $d_p = 2$ mm; $H_o = 325$ mm) is shown in [Figure 18](#). For $z/D = 1.1$, the deviation between the profiles increase by 8.32%, 10.13%, and 22.13% with increasing the U/U_{ms} ratio from 1.1 to 1.2, 1.3, and 1.4, respectively. However, the deviation between the profiles decreases with increasing the

z/D levels and becomes less at $z/D = 1.8$. The deviations between the profiles at $z/D = 1.8$ were 10.57%, 15.52% and 23.96% for U/U_{ms} equals to 1.2, 1.3, and 1.4, respectively. For each U/U_{ms} ratio, the gas holdup decreases with the increase in the bed height and decreases radially towards the spout-annulus interface, where the maximum gas holdup was obtained at the axis of the spout. The results clearly show the effect of the U/U_{ms} ratio on the gas holdup in the spout. Increasing the U/U_{ms} causes the gas holdup values to increase substantially in the entire spout. This implies that applying spouted beds at higher U/U_{ms} ratios, even when these ratios are within stable range of operation, will cause the spout to become bigger and more dilute (see [Figure 15](#) and [Figure 16](#)). Thus, particles in the spout will have less chance to contact with the reactive gas due the violent spouting caused by the large energy transferred from the gas.

4. Conclusion and perspectives

Gamma-ray computed tomography has been successfully used to determine the gas-solid cross-sectional distributions of spouted beds. The quality of the TRISO nuclear fuel particles is strongly impacted by the hydrodynamics, flow field, and flow regime characteristics of the spouted beds. The gas-solid distributions of spouted beds is important from the TRISO fuel coating perspective, as the deposition of the coating layers on the particles depend on how and where the particles are present in the bed, which, in turn, is impacted by the design parameters and operating variables. Experiments that studied the effect of particle density and size, and bed size on the solids holdup distributions were performed at different operating conditions. The results lead to further discussion about the influence of the U/U_{ms} on the gas and solids holdups distributions of spouted beds. On the basis of the results obtained, some of the important conclusions are the following:

- The results of the solids holdup of particles of different densities and sizes revealed that the assumption that stable spouting mode would result in uniform coating layers for the particles in the TRISO fuel coating process is not adequate. The results showed that even though the particles were operated at stable spouting state and same U/U_{ms} and H_o , differences were obtained in the solids holdup distributions in the spout and the fountain regions. Hence, the changes in the properties of the particles must be accompanied by appropriate changes in the input process of the spouting conditions to produce particles with uniform coating layers. This finding needs to be considered for the practical process of the TRISO fuel coating.
- It has been found that the gas holdup values in the annulus are higher than the mean gas holdup value at the loose-packed bed state when the gas flow rate increases much the U/U_{ms} . The common assumption that the gas holdup in annulus is equal to the loose-packed bed value is not proven, especially with increasing the gas flow rate. Therefore, findings, measurements techniques, CFD models, and the reported closures for simulating spouted beds based on this assumption can be challenged due to accuracy.
- The originality of the results and analysis presented in this contribution provides the basis for the assessment of the design of the TRISO fuel coater and its modelling to achieve the proper distribution of the phases and to obtain uniform coating layers.
- At last, successful implementation of the CT as a non-invasive technique on the cold-flow coater will lay the ground for its implementation on hot pilot plant and industrial units.

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Notation

d_p	particle diameter, mm
D, D_c	inner column diameter, m
D_i	inlet orifice diameter, m
H_o	static bed height, mm
L	column height, m
R	column radius, cm
T	bed temperature, °C
U	superficial gas velocity, m s ⁻¹
U_{ms}	minimum spouting velocity, m s ⁻¹
z	axial distance form inlet orifice, m

Greek letters

ρ_p	particle density, g cm ⁻³ .
ϕ_p	sphericity of particles.
ε_o	gas holdup (voidage) at packed bed state
γ	cone angle, ° degree

5. References

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Table 1. Properties of the materials used.

Material	d_p (mm)	ρ_p (g/cm ³)	ϕ_p	ε_o
Glass beads	1	2.4	1	0.413
	2	2.5	1	0.411
Steel particles	1	7.4	0.98	0.412
	2	7.4	0.98	0.411

Gas-solid Spouted bed

Gamma-ray computed tomography (CT) and spouted bed inside the setup for scan.

Cross-sectional distributions of solids holdup for gas-solid spouted bed.

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Figure 1

Figure 2

Figure 4

Figure 7

Figure 8
(a)

Figure 9

Figure 10

Figure 13

Figure 14

Figure 15

Figure 16

Figure 17**(a) $z/D=0.8$** **(b) $z/D=1.1$** **(c) $z/D=1.8$**

Figure 19