Original Article

Disposal of Uranium and Thorium Entering the Human Digestive System before Reaching the Blood by Adsorption on Simethicone and Forming a Non-absorbable Compound: A Full *in vitro* Analysis

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ABSTRACT

Introduction: This method was applied to form a non-absorbable complex with uranium and thorium swallowed with contaminated dust or other sources to get rid of them before absorption into the blood and discarded outside the body. A non-GIT absorbable simethicone was used in this method for the adsorption of uranium (VI) and thorium (IV) from a solution similar to the gastrointestinal tract pH medium. **Materials and Methods:** Accurately measured 9 mg of pure standard uranyl nitrate and simethicone emulsion in concentration range (100-300 mg) at 37 ± 1 °C for 20 min in pH ranges like the medium of the stomach and intestine then take the supernatant after centrifugation to make sure of decreasing of the concentration of uranium and thorium ions by ICP Mass Spectroscopy (ICP-MS). **Results and Discussion:** The data recorded for different total uranium and thorium concentrations reveal convergent behavior. Increasing simethicone concentration inside the safe dose shifts the sorption of uranium and thorium to 100 % of the total concentration, calculated t and F values were compared with the reference method and the proposed method was equally precise and accurate. **Conclusion:** The data indicate that the method can successfully be applied for the complete adsorption of uranium and thorium on simethicone from GIT before reaching the blood.

Key words: Simethicone, Uranium, Thorium, Protection, Non-absorbable complex.

INTRODUCTION

Uranium and thorium are heavy metals with chemical toxicity and radioactivity, which causes progressive or irreversible renal injury and their compounds are potential carcinogens.[1,2]

Simethicone is a chemically inert drug that is not absorbed from the gastrointestinal tract[3,4] used to relieve the symptoms of excessive gases in GIT.[5,6]

Recently, a new type of inorganic– organic composite chelating adsorbent had been synthesized to selective laboratory adsorption. The composite adsorbents always contain a chelating agent and an inorganic particle matrix such as silica,[7] smectite[8] and bentonite.[9]

Uranium dust content varies according to the type of the source,[10-12] it was determined by complex formation using piroxicam,[13] meloxicam,[14] 2-(2- thiazolylazo)-p-cresol (tac),[15] arsenazo iii,[16] 2-ethanolimino-2-pentylidino-4-one,[17] a mixture of - xylene and benzene,[18] azide ions,[19] acetylacetonate.[20]

Many procedures have been developed for determination of thorium including liquid–liquid extraction,[21,22] ion-exchange resins,[23,24] liquid membrane[25] and solid phase extraction (SPE).[26-29]

According to U.S. Food and Drug Administration approval of simethicone, we were select it due to the safety on the human body and not need more tests on humans.[30]

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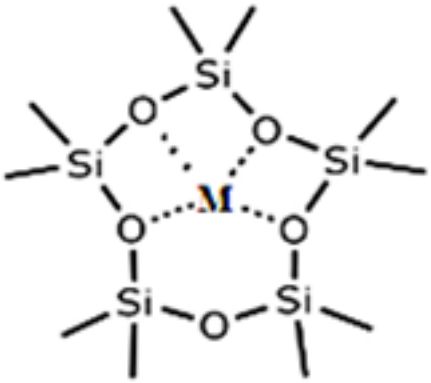
Dust contaminated with uranium and thorium is one of the sources of the exposure of the human body to these radioactive materials by inhalation or ingestion.[31,32] If uranium and thorium reached the blood it will be difficult to remove and will cause many diseases and physical problems.[33,34]

The main purpose of the present study is to find a way to prevent the absorption of uranium and thorium from the digestive tract to the blood and take them out of the body, simethicone was used for removal of uranium (VI) and thorium (IV) from GIT medium. The influences of time, pH, amount of adsorbent and competitive substances on the sorption were investigated in details. The formed chelate may be due to the following mechanism (Scheme 1).

MATERIALS AND METHODS

Materials and reagents working solution

Chemicals used for the method were of the highest purity as available from their sources in the form of pure analytical grade.



Scheme 1: A suggested structural mechanism of the formed chelation complex between simethicone and M (thorium or uranium)

1-Uranyl nitrate: 4 mgml^{-1} solution in distilled water obtained from Fluka/Sigma-Aldrich (St. Louis, MO).

2-Thorium nitrate: 4 mgml^{-1} solution in distilled water obtained from Sigma Chemical Co. (St. Louis, MO, USA)

3-Simethicone emulsion 2%, USP 29, Amriya Pharm Industries.

Methods

Accurately measured 9 mg of pure standard uranyl nitrate and thorium nitrate were transferred into a 25 ml volumetric flasks then add simethicone emulsion in concentration range (100 -300 mg), the volumes were completed with distilled water and then immersed in a water bath at 37 ± 1°C for 20 min for each pH from (1.5 to 3.5) like the medium of the stomach and from (5.5 – 7.5) similar to the intestine medium with continuous stirring and the volume was adjusted with double distilled water to 25 ml. For each sample, the supernatant was separated from the adsorbent by centrifugation at 3000 xg for 5 min, then we took the supernatant liquid to make sure decreasing of the concentration of uranium and thorium ions by ICP Mass Spectroscopy (ICP-MS) at pH 1.5 - 7.5. The interference effect of pepsin, pancreatin, fatty acids (palmitic and oleic acid), amino acids (glycine, glutamine and arginine), monosaccharide's, lipase, proteases and amylases on this method had also been studied, uranium and thorium are a primary unit that will not be digested, as well as simethicone is not affected by digestive juices and is not absorbed.[8,9] The proposed method was equally precise and accurate as of the reference one according to statistical analysis of the results, calculated t and F values were compared with the reference method[35] to assessing the performance of the proposed method and showing no significant difference between them.

Table 1: Back (ICP-MS) analysis of standard uranium (360 μg ml^{-1}) adsorbed on simethicone emulsion in concentration range (100 -300 mg) at different pH 1.5 – 7.5 in supernatant solution.

Uranium	Simethicone emulsion	pH					
		1.5	2.5	3.5	5.5	6.5	7.5
Taken ppm	Taken mg	Uranium remaining in supernatant (ppm)					
360	100	332	335	338	225	210	202
360	150	167	167	165	160	145	135
360	200	102	105	100	120	75	70
360	250	38	36	35	33	10	9
360	300	0	0	0	0	0	0

RESULTS AND DISCUSSION

The previous procedure was applied to:

a- Insurance of complete adsorbance of uranium and thorium by back (ICP-MS) analysis of high concentrations of pure standard uranium and thorium (360 μg ml^{-1}) adsorbed on simethicone emulsion in concentration range (100 -300 mg) at different pH 1.5 – 7.5 (Tables 1,2), the results obtained compared with the reference method[35] are shown in (Tables 3,4).

b- 360 μg ml^{-1} of uranium and thorium in presence of 10 μg ml^{-1} oleic acid, amino acids (glycine, glutamine and arginine), monosaccharide's, Lipases, Proteases and amylases, the results obtained are shown in (Tables 5,6). The experimental back (ICP-MS) determination of remaining uranium and thorium in the supernatant at different pH values indicate the complete adsorption of them on simethicone. The data recorded for different total uranium and thorium concentrations

Table 2: Back (ICP-MS) analysis of standard thorium (360 μg ml^{-1}) adsorbed on simethicone emulsion in concentration (100 -300 mg) at different pH 1.5 – 7.5 in supernatant solution.

Thorium	Simethicone emulsion	pH					
		1.5	2.5	3.5	5.5	6.5	7.5
Taken ppm	Taken mg	Thorium remaining in supernatant (ppm)					
360	100	300	331	340	220	215	198
360	150	164	169	168	157	152	135
360	200	100	109	102	115	78	69
360	250	35	30	38	34	15	6
360	300	0	0	0	0	0	0

Table 3: Statistical data for the adsorption of uranium on simethicone, compared with the reference method.

Statistic	pH					
	1.5	2.5	3.5	5.5	6.5	7.5
Mean recovery*± S.D.	99.44± 1.56	99.68±0.87	99.04±0.717	99.7±0.717	100.34±0.82	99.82±0.63
N	5	5	5	5	5	5
Variance	2.423	0.757	0.514	0.515	0.668	0.407
S.E	0.695	0.389	0.32077	0.320	0.365	0.2853
t-test	0.35	0.7	1.045	0.27	2.32	0.04
F-test	6.3	1.83	1.999	1.05	1.28	2.00
Range of recovery (%)	99.04 - 100.34					

Table 4: Statistical data for the adsorption of thorium nitrate on simethicon compared with the reference method.

Statistic	pH					
	1.5	2.5	3.5	5.5	6.5	7.5
Mean recovery*± S.D.	100± 0.725	99.62±1.013	100.24± 0.73	99.6± 0.839	98.98±0.83	99.18±0.559
N	5	5	5	5	5	5
Variance	0.525	0.712	0.533	0.705	0.697	0.312
S.E	0.324	0.377	0.326	0.375	0.373	0.2498
t-test	2.2	0.17	1.98	0.8	1.64	1.648
F-test	1.5	1.44	1.02	1.36	1.17	4.433
Range of recovery (%)	98.98 - 100.24					

(*) Average of three experiments

Table 5: Effect of some common ingredient in GIT, on the adsorption of uranium on simethicone.

Other Ingredients 10 µg. added	Uranium		
	Taken ppm	Recovery* %	pH
—	360	99	2.5
pepsin	360	98.7	2.5
pancreatin	360	99.8	2.5
fatty acids	360	101.5	7.5
amino acids	360	99.1	7.5
monosaccharides	360	99	7.5
Lipases	360	98.9	7.5
Proteases	360	99.3	7.5
Amylases	360	99.9	7.5

.(*) Average of three experiments

Table 6: Effect of some common ingredient in GIT, on the adsorption of thorium on simethicone.

Other Ingredients 10 µg. added	Thorium		
	Taken ppm	Recovery* %	pH
—	360	99.2	2.5
pepsin	360	99.7	2.5
pancreatin	360	98.8	2.5
fatty acids	360	100.3	7.5
amino acids	360	98.1	7.5
monosaccharides	360	98	7.5
Lipases	360	99.7	7.5
Proteases	360	99.2	7.5
Amylases	360	99.6	7.5

.(*) Average of three experiments

reveal convergent behavior. Increasing simethicone concentration inside the safe dose shifts the sorption of uranium and thorium to 100 % of the total concentration (discussed further on). Simethicone drug was chosen as it is safe, non-GIT absorbed, very popular and low cost. The procedure is very simple, fast and the dose used for protection far from the maximum allowable dose (500 mg simethicone).

Therefore, according to the average time for digested food to start coming out the stomach (1-2) hr,[36] we recommend that the worker exposed to contaminated dust take a half teaspoon(≈2.5ml) of Simethicone 2% (≈50mg) just before starting to exposure to contaminated dust and after 1 hr of exposure. If the number of exposure hours exceeds, a half teaspoon is taken every 1 hr with a maximum 10 doses per day within 10 working hours in order to not exceed the allowable dose/day (500mg). The optimization of sorption parameters was carefully studied depending on the duration time of uranium and thorium in the parts of the digestive system and the required degree of acidity as mentioned in the following paragraphs:

Effect of heating temperatures

GIT similar temperature (37 ± 1°C) was sufficient to achieve the goal of the complete adsorption of uranium and thorium on simethicone. Many temperatures were tried, the optimum temperature was from 25 - 50°C for uranium and 25 – 45°C for thorium at pH 5.5 and 7.5, (Figures 1 and 2).

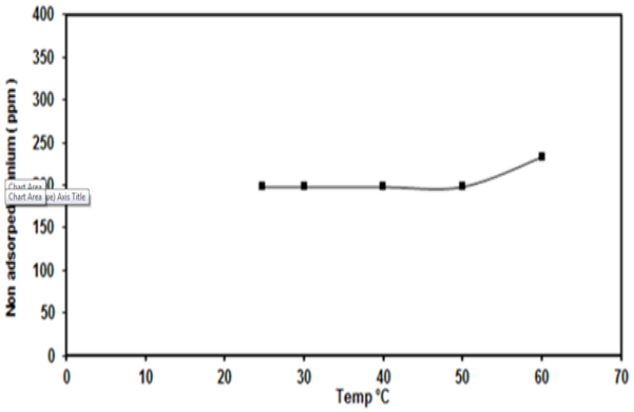


Figure 1: Effect of temperature on the adsorption of 360ppm uranium on 100mg simethocone at pH 5.5 and 7.5.

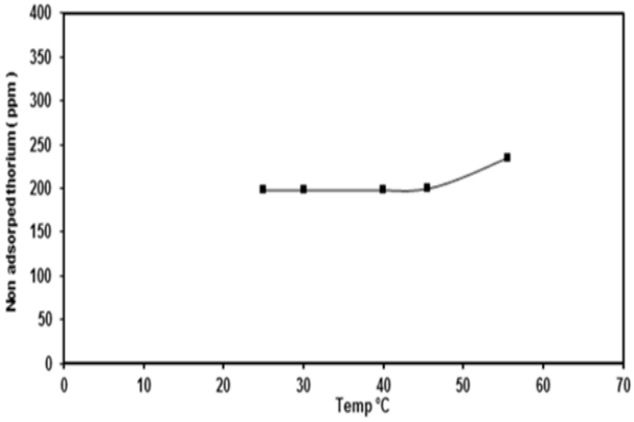


Figure 2: Effect of temperature on the adsorption of 360ppm thorium on 100 mg siethicone at pH 5.5 and 7.5.

Effect of time

The reaction was carried out at 37 ± 1°C and the adsorption complex formed was stable at 20 min to 17 hr for uranium and 15 hr for thorium at pH 5.5 (Figure 3), but at pH 7.5 the complex was stable for more than 72 hr for uranium and thorium (Figure 4).

Effect of simethicone concentration

It was found that 100 mg (1 teaspoon) of 2% simethicone achieves a suitable concentration for the adsorption of uranium and thorium in the range of (22-58 ppm) and (20-62 ppm) respectively along with the GIT parts, at very high concentration of uranium and thorium (360 ppm) the 300mg (3 teaspoons or 1 tablespoon) of 2% simethicone were sufficient for full adsorption at different pH, (Figures 5,6).

Effect of pH

pH was carefully examined by studying the solutions similar to the gastrointestinal tract pH medium adjusted by adding 5% v/v of HNO3 or 5% of v/v NH_4OH solutions between 1.5 to 7.5, the results showed that, the adsorption efficiency slightly different with the change in pH. At pH 1.5, adsorption starts to slightly decreases but increases again at pH 5.5, sorption still slightly increases to pH 7.5, pH 1.5 to 7.5 can be selected as optimum pH for uranium and thorium sorption on simethicone in GIT, (Figures 7,8) show the variation of the adsorption as a function of pH values.

Interference Study

To study the effect of GIT enzymes and digested products as pepsin, pancreatin, lipase amylases, proteases, fatty acids (palmitic and oleic acid), amino acids (glycine, glutamine and arginine) and monosaccharide's on the adsorption of uranium and thorium, The test was applied to each substance on its acidic medium, the data demonstrate that no effect of this compound on expelling these dangerous metals from the digestive tract.

Linearity and quantification

An inverse linear relationship was obtained in back (ICP-MS) measurement between simethicone concentration and supernatant concentration of remaining uranium and thorium (Figures 5,6).

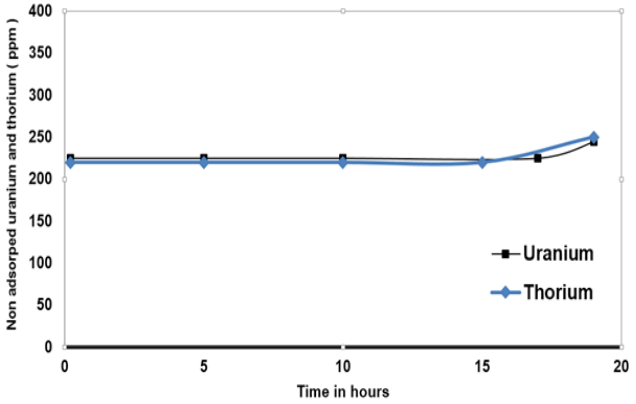


Figure 3: Effect of time on the adsorption of 360ppm uranium and thorium on 100 mg simethicone at pH 5.5

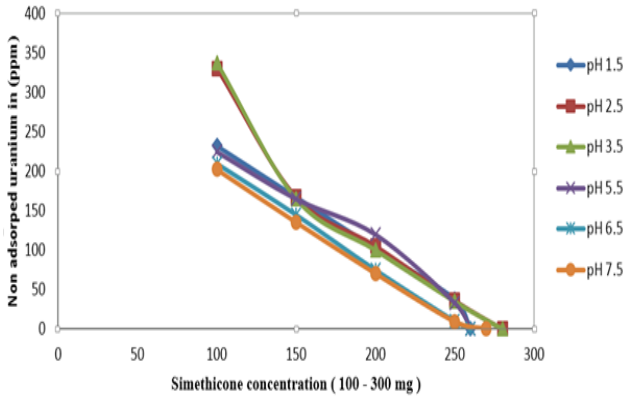


Figure 5: Effect of simethicone concentration (100 – 300 mg) on the adsorption of uranium at different pH (1.5. – 7.5).

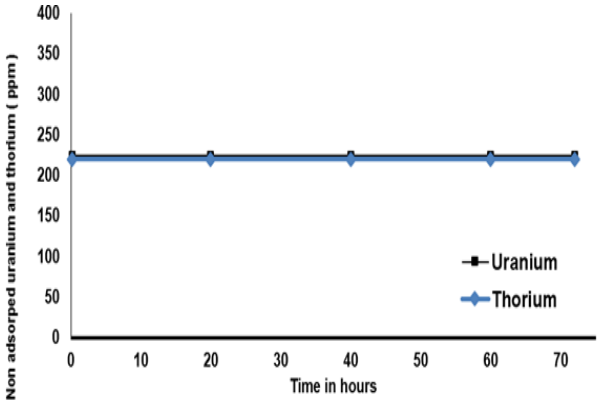


Figure 4: Effect of time on the adsorption of 360ppm uranium and thorium on 100 mg simethicone at pH 5.5.

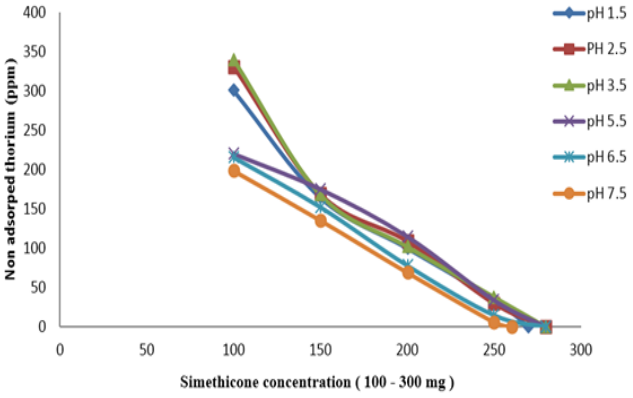


Figure 6: Effect of simethicone concentration (100 – 300 mg) on the adsorption of thorium at different pH (1.5. – 7.5).

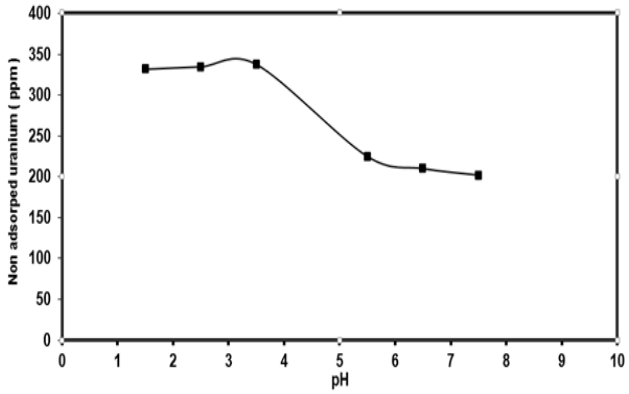


Figure 7: Effect of pH on the adsorption of 360ppm uranium on 100 mg simethicone.

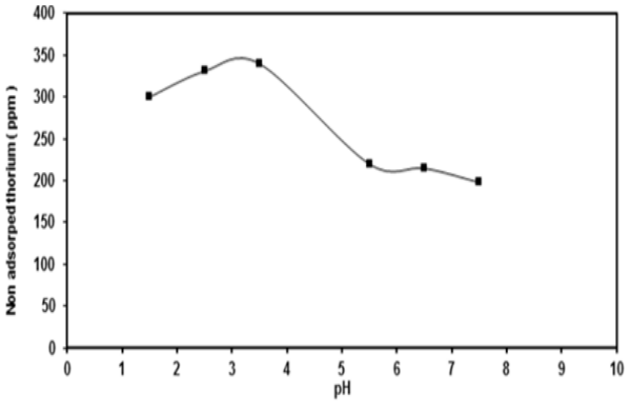


Figure 8: Effect of pH on the adsorption of 360ppm Thorium on 100 mg simethicone.

The proposed method was equally precise and accurate as of the reference one according to the statistical analysis of the results of the three replicate determination values under the same conditions for each sample analysis. Accuracy and precision were indicated by calculating t and F values which were less than the tabulated ones indicating that there was no significant difference between the proposed and reference method[35] (Tables 3,4). The small SE indicate excellent precision and accuracy, recovery experiments were performed by adding a known amount of the enzymes and digestion units of food. No interference from the common additives was observed in the results (Tables 5,6) as an additional confirmation of accuracy and precision. The good linearity of the calibration graph and the negligible scatter of the experimental points were clearly evident from the value of the variance. Therefore, the method is useful for the disposal of uranium and thorium entering the human digestive system before reaching the blood.

CONCLUSION

The proposed method was successfully applied for the complete adsorption of uranium and thorium on simethicone from GIT before reaching the blood, The method is successful in the various acidic medium along the digestive tract, The data was assessed by comparing the calculated t and F values with the reference method[35] Tables 3,4, the results showed no significant difference between them. In (Tables 5,6) the results obtained show that pepsin, pancreatin, lipase, amylases, proteases, fatty acids (palmitic and oleic acid), amino acids (glycine, glutamine and arginine) and monosaccharide's do not interfere. Statistical analysis of the results revealed that the proposed method was more precise and accurate than the reference one.

ACKNOWLEDGEMENT

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CONFLICT OF INTEREST

The authors report no conflicts of interest.

ABBREVIATIONS

GIT: Gastro Intestinal Tract; **ICP-MS:** ICP Mass Spectroscopy.

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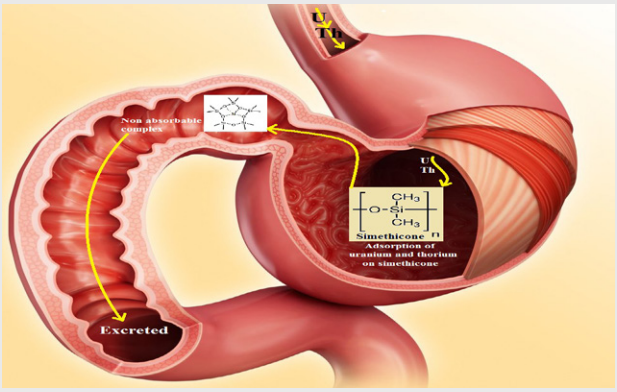
PICTORIAL ABSTRACT



SUMMARY

The method is based on protecting the body of the person who is exposed to inhalation or swallowing the uranium and thorium dust, this is done by preventing the absorption of these dangerous elements into the blood by using simethicone as a safe drug to make a non-absorbable complex that is excreted outside the body.

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