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Measurements of the density of radioactive particles using sodium polytungstate

ABSTRACT

Thirteen MTR-type particles and four DFR-type particles were measured for density using a method based on their behaviour in sodium polytungstate solutions at different densities. Estimates of particle density were assigned by observing if the particle sank or floated in the solutions prepared. The method was calibrated using aluminium of certified density.

The results showed a density range of about 2.7 to 3.1 g/cc for MTR particles, similar to those predicted from knowledge of fuel composition and uranium content. In the case of DFR particles, the apparent densities were all greater than about 3.1 g/cc, probably reflecting the heavy main elemental constituents U, Nb and Fe.

There were some observable differences in MTR particle densities depending on where they were found (seabed, Dounreay foreshore, Sandside beach), but the limited number of results precludes full assessment of the significance of these differences.

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1. INTRODUCTION

The measurement of density of small radioactive irradiated fuel fragments (hereinafter called particles) which are found in the local environment at Dounreay, has proved to be problematic. A review of possible techniques which could be used was reviewed by RM Consultants Ltd [1]. Further investigations of potentially viable techniques e.g. particle settling in water column and gas pycnometry showed that they would not deliver reliable measurements. The use of photomicrographs of particles, taken in plan view and from various angles, coupled with computer aided design software for assessing particle volume will provide density estimates if the mass is first measured. This technique was tried on ten MTR-type particles and provided densities in the range of 1.7 to 3.3 [2]. The lower results were thought to be possibly due to the presence of voids or re-entrant areas on irregularly shaped particles, which the software could not cope with.

Based on the known uranium content of the MTR fuel alloys used and on tabulated data of densities for U/Al fuel [3], the expected range of densities is from about 3.1 to 3.4. This would be extended down to nearly 2.7 depending on the amount of aluminium cladding attached to the particle. Figure 1 shows a fuel plate consisting of uranium-aluminium fuel sandwiched between aluminium cladding. When such fuel plates were being milled under water, particles (swarf) would have been generated which could consist of aluminium cladding only, fuel only or any combination of the two.

It was decided to try another method for density measurement, this one based on preparation of sodium polytungstate solutions at different densities. Estimates of density were assigned by observing if the particle sank or floated in the solutions prepared. The method was calibrated using aluminium of certified density.

2. METHODOLOGY

A series of aqueous solutions of sodium polytungstate (SPT) were prepared to provide solutions of density 2.6, 2.8 and 3.0 approximately. The particle of interest was placed in the solution and the sample centrifuged. The particle will either rise to the top of the solution and float, or will sink to the bottom. From this, the density of the particle can be determined as either less than or greater than the measured density of the solution. This process can be repeated in solutions of different density and the particle density can be assigned to lie in a certain limited range. The method is described more fully elsewhere [4].

3. RESULTS

In total, thirteen MTR particles and four DFR particles were measured by the SPT technique. The results are presented in Table 1 and are presented graphically in Figure 2. The results showed a density range of about 2.7 to 3.1 g/cc for MTR particles, similar to those predicted from knowledge of fuel composition and uranium content.

In the case of DFR particles, the densities measured were all greater than about 3.1 g/cc, probably reflecting the relatively high density of the main elemental constituents U, Nb and Fe. Although DFR particles have a porous structure, it appears that, at least for the samples tested, this porosity is insufficient to reduce the density below 3.1. It is recognised that the presence of internal pores in these DFR particles may have had an influence on the measured density; therefore, the measurements for the four DFR particles may be termed ‘apparent densities’.

Stable solutions of sodium polytungstate have an upper limit at a density of about 3.1 g/cc. Solution densities higher than this, and potentially up to 4.0 g/cc, could be prepared by the addition of tungsten carbide (WC) powder. The WC does not dissolve in the SPT solution but instead forms a solid suspension which would separate if centrifuged. Attempts to increase the density of the liquids above 3.1 have not been made - there are some concerns about being able to easily retrieve small particles from the WC-SPT mixture and the timescales involved in gravity settling could be very long.

There were some observable differences in MTR particle densities depending on where they were found (seabed, Dounreay foreshore, Sandside beach), but the limited number of results precludes useful assessment of the significance of these differences.

Intuitively, one might expect a positive correlation between density and ^{137}Cs activity - the more uranium in the fuel fragment then the more fission products would be present. Indeed there is a suggestion of a positive slope in Figure 2. However, there are confounding factors in that (1) there may be different amounts of inactive aluminium associated with a fuel fragment and (2) the amount of fission of the ^{235}U present will depend on irradiation history - different fuel fragments may have been irradiated at different neutron fluxes, at different positions in the core or for different times.

A comparison of results for four MTR samples which have been analysed by two methods shows quite poor agreement – see Figure 3. One of the two particles where the CAD method showed a lower result than the SPT method is peculiarly shaped (941508) although the other one is fairly regular (L80PART/05/008) - see UKAEA report EPD(06)P255 [2]. It is considered that the present SPT technique is superior to the CAD method and any additional measurements should make use of this method.

4. REFERENCES

[1] Black, J (2004). Study of techniques for measuring small particle densities. RMC Report R04-071(T).

[2] Toole, J (2006). Estimation of MTR particle density using photomicrographs and volume calculation by CAD software. UKAEA Report EPD(06)P255.

[3] Kaufman, A.R. (Editor) (1962). Nuclear reactor fuel elements – metallurgy and fabrication. Interscience Publishers.

[4] MacGregor, J (2007). Particle density measurement – inactive trials. UKAEA Chemistry Support Services, Note for the record CSS/NFR(07)P09.

Table 1 - Results of Density Measurements for MTR and DFR Particles

Particle	Type	Location	Behaviour in prepared sodium polytungstate solution after centrifugation			Conclusion	Comments
				2.908g/ml	**2.585g/ml**		
Certified Aluminium				floats	sinks	Density >2.585, <2.908	Aluminium with a known density of 2.70 g/ml was used as a method performance check

Particle	Type	Location	3.084g/ml	2.861g/ml	2.554g/ml	Conclusion	Comments
941508	MTR	Foreshore	floats	suspended	not tested	Density close to 2.861	
L80PART/05/009	MTR	Foreshore	sinks	not tested	not tested	Density > 3.084	
L80PART/03/104	MTR	Foreshore	floats	floats	sinks	Density > 2.554, < 2.861	
L80PART/06/044	MTR	Sandside	floats	floats	sinks	Density > 2.554, < 2.861	The particle was observed to sink very slowly, suggesting a density only slightly higher than 2.554.
L80PART/07/006	MTR	Sandside	floats	floats	sinks	Density > 2.554, < 2.861	The particle was observed to sink very slowly, suggesting a density only slightly higher than 2.554.
L80PART/07/024	MTR	Sandside	floats	suspended	not tested	Density close to 2.861	
L80PART/06/035	DFR	Sandside	sinks	not tested	not tested	Density > 3.084	
L80PART/05/053	MTR	Seabed	floats	floats	sinks	Density > 2.554, < 2.861	
L80PART/05/064	MTR	Seabed	floats	floats	sinks	Density > 2.554, < 2.861	

Particle	Type	Location	3.091g/ml	2.877g/ml	2.568g/ml	Conclusion	Comments
L80PART/05/008	MTR	Foreshore	sinks	not tested	not tested	Density >3.091	As this result was very different from the photomicrography-CAD method, it was repeated to confirm
L80PART/05/008	MTR	Foreshore	sinks	not tested	not tested	Density >3.091	
L80PART/05/130	DFR	Seabed	sinks	not tested	not tested	Density >3.091	The particle was observed to sink very slowly, suggesting a density only slightly higher than 3.091.
L80PART/06/042	MTR	Foreshore	floats	suspended	not tested	Density close to 2.877	
Certified Aluminium			floats	floats	sinks	Density >2.568, <2.877	Aluminium with a known density of 2.70 g/ml was used as a method performance check

Particle	Type	Location	3.075g/ml	2.810g/ml	2.538g/ml	Conclusion	Comments
Certified Aluminium			floats	floats	sinks	Density >2.538, <2.810	Aluminium with a known density of 2.70 g/ml was used as a method performance check
L80PART/04/119	MTR	Seabed	floats	suspended	not tested	Density close to 2.810	
L80PART/04/102	MTR	Seabed	not tested	floats	sinks	Density >2.538, <2.810	
L80PART/05/054	MTR	Seabed	floats	sinks	not tested	Density >2.810, <3.075	

Particle	Type	Location	3.049g/ml	2.806g/ml	2.529g/ml	Conclusion	Comments
L80PART/06/039	DFR	Sandside	sinks	not tested	not tested	Density >3.049	
L80PART/05/138	DFR	Seabed	sinks	not tested	not tested	Density >3.049	
Certified Aluminium			floats	floats	sinks	Density >2.529, <2.806	Aluminium with a known density of 2.70 g/ml was used as a method performance check

L80PART/05/094	MTR	Seabed	NA	NA	NA	NA	Particle was stuck in putty on microscope stub and could not be cleanly removed
L80PART/07/012	DFR	Sandside	NA	NA	NA	NA	Particle was physically too small to analyse

These particles were analysed on different days and the densities of the sodium polytungstate solutions were re-measured.

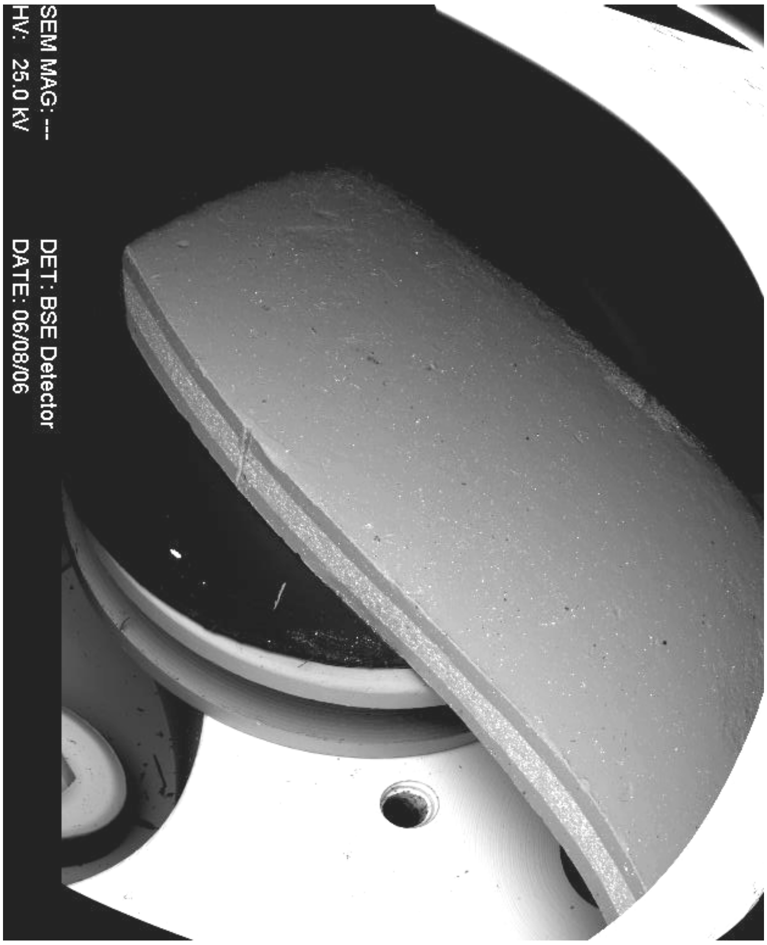

Figure 1 - Example of an unirradiated MTR fuel plate. Depth of the plate is approximately 2 mm.

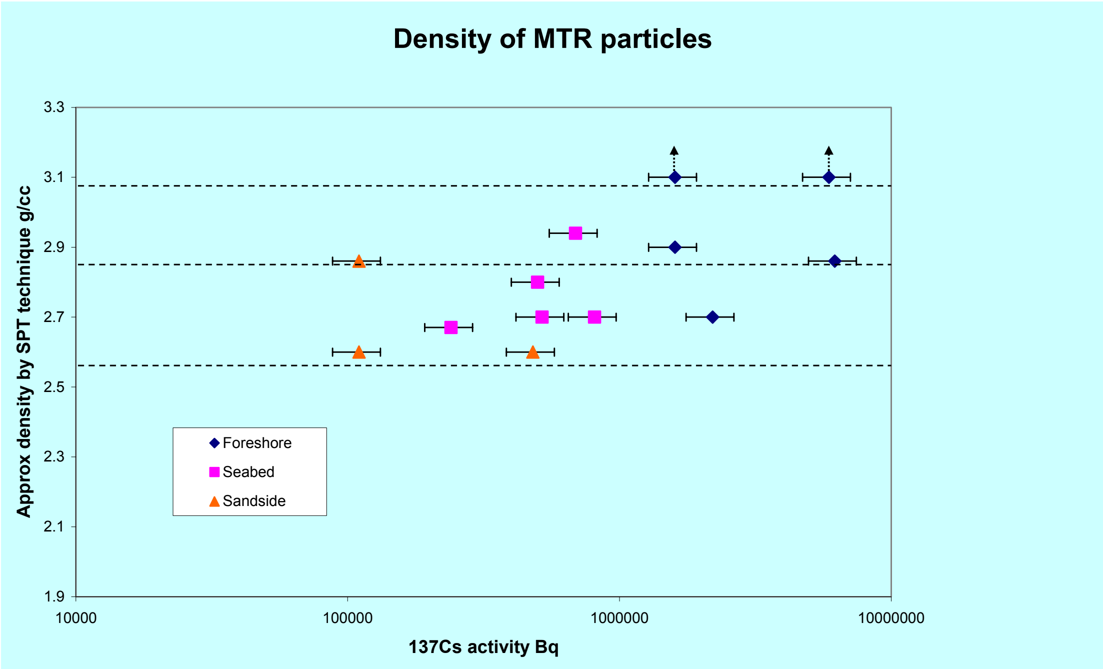

Figure 2 - Measured densities of MTR particles, arranged by location and ^{137}Cs activity

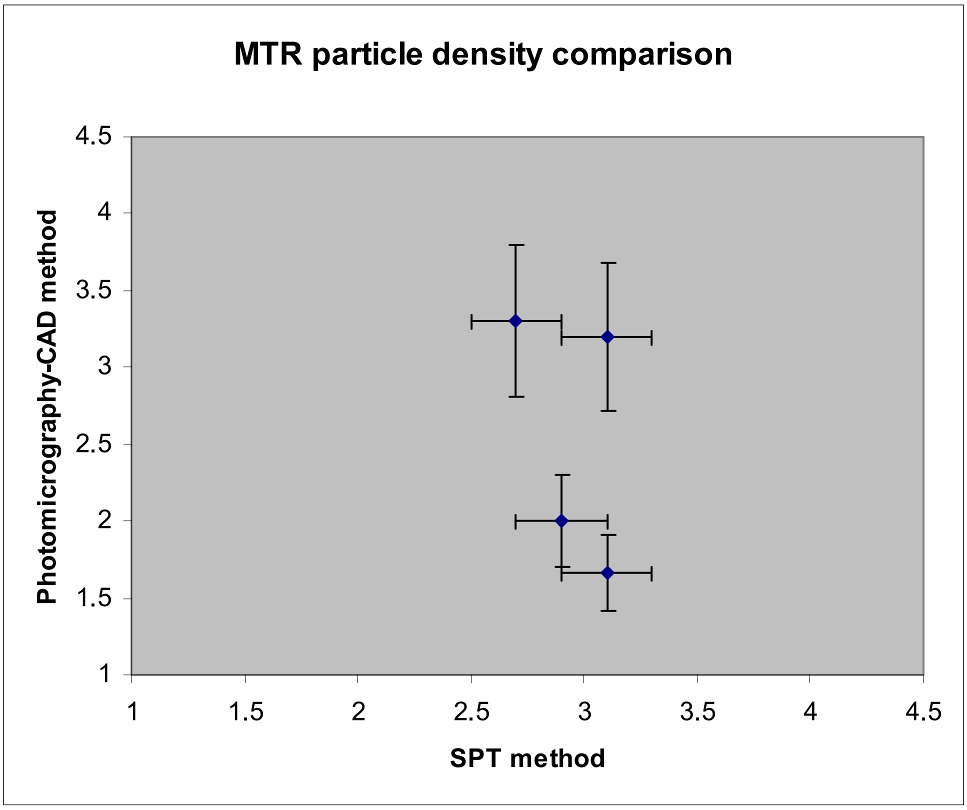

Figure 3 - A comparison of MTR particle density results for two independent techniques