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Hot Particles in the Terrestrial Food Chain: Sources, Characterisation and Radiological Implications

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1. Introduction

A hot particle in this study is defined as a particle of solid material that emits radiation in the form of alpha particles at a rate far greater than that of neighbouring material. The size of the particle cannot be defined but sizes reported previously are of the range 5 - 500 μm diameter. The alpha activity associated with hot particles can range from mBq to Bq (beta and gamma activities can be as high as several MBq in the case of Chernobyl hot particles). Present scientific convention has adopted the use of the word 'hot' in this field of research, which can be evocative and misleading in some cases. Certainly in this study, low levels of alpha emissions can be detected against background conditions, but the levels of activity would not be considered 'hot' in other fields of radiation research. Therefore there is the need for the classification of hot particles with respect to size, total activity, radiation type, source and composition.

Hot particles may be of natural origin. In igneous rocks around 90% of the uranium and thorium is concentrated in the accessory minerals, such as apatite, fluorite, monazite, sphene or zircon, which usually constitute less than 1% of the volume of the rocks (Hamilton 1988). Weathering of the parent rock releases these dense, abrasion resistant minerals, which are then concentrated in areas of high energy, such as rivers and coastal regions. Accessory minerals typically have mean particle diameters of less than 50 μm.

Of potentially greater radiological significance are artificial hot particles. These were first introduced into the environment in the atomic detonations of 1945 (Adams *et al.* 1960) and continued to be released throughout the period of atmospheric nuclear weapons testing. In the UK, between 1951 and 1957, routine operations at the Windscale plant led to the release of radioactive particles into its immediate vicinity (Chamberlain 1991). A number of accidents at nuclear installations have also led to the release and dispersal of hot particles, most notably the Chernobyl disaster in 1986. Most of the activity within 30 km of the power plant is associated with hot particles (Askbrant *et al.* 1996), and particles from Chernobyl have been discovered as far afield as Scandinavia (Raunemaa *et al.* 1987), Germany (Rudhard *et al.* 1992) and Bulgaria (Balashazy *et al.* 1988). Other incidents responsible for the dispersal of radioactive particles include the Windscale fire of 1957, the explosion at Kyshtym reprocessing plant in 1957, and satellite break-up on re-entry to the atmosphere.

Radioactive particles have occasionally been detected and isolated in the vicinity of Sellafield. McMahon *et al.* (1994) describe irradiated uranium oxide particles collected from the Windscale area in 1956. After the 1957 fire, which resulted in the shutting down of the Windscale Piles, district survey teams found and collected large numbers of active particles (Dunster *et al.* 1958). Day *et al.* (1992) have obtained evidence for uranium oxide particles in woodland close to Sellafield, and more recently Evans (pers. comm.) has located radioactive particles within the Sellafield perimeter.

During the 1980's some attention was given to the presence of particulate activity in the UK marine environment as hot particles were found to be present in low-level liquid waste discharged to the coastal marine environment (Hamilton & Clarke 1984; Cook *et al.* 1984; Hamilton 1985; Kershaw *et al.* 1986; Baxter *et al.* 1989). Recent discoveries of environmental contamination in the Dounreay area (New Scientist 1995; ENDS Report 1995) have also drawn attention to the problem of very active hot particles on the Caithness shoreline.

In the terrestrial environment, atmospheric discharges from the nuclear industry are the principal source of hot particles. The aerodynamics of the particulate material, its density, the height of release and the prevailing weather conditions govern its range of travel.

The role that hot particles may play in radiation dose delivery to members of the UK public through the ingestion of terrestrial foodstuffs is unclear. Indeed the dose delivered by hot particles has been the subject of considerable debate for many years. Tamplin and Cochran (1974) hypothesised that the risk of cancer induction for a given dose from particulate plutonium in the lung could be five orders of magnitude higher than the risk predicted from the mean organ dose. Subsequent research has refuted their argument, and it is now widely accepted that the carcinogenicity of a localised dose is actually considerably less than that of a uniform dose (Hofmann *et al.* 1989; Charles 1991). Even so, information is still required regarding the abundance of hot particles in UK terrestrial foodstuffs.

Hot particles have been well studied in the UK marine environment and in the vicinity of Chernobyl, with regard to characterisation, sources and dose assessments. This project,

however, specifically addresses the situation of whether or not hot particles are present in UK terrestrial foodstuffs and if so, what are the radiological implications.

2. Aims and Objectives

The aims of this project, as agreed with MAFF, are to:

- provide a critical review of the scientific literature regarding the emissions and properties of hot particles from routine operations of nuclear installations as well as those emitted during the Chernobyl disaster;
- quantify the abundance of hot particles in the terrestrial environment in the vicinity of Sellafield;
- characterise such particles with regard to their elemental, mineralogical and radionuclide composition;
- assess the availability of hot particles to the food chain, either by inhalation or ingestion; and
- radiologically assess the hot particle content of a number of food items.

A literature review was undertaken and submitted in early August 1995 - MAFF Report 1B078 (Whittall & McDonald 1995).

Following several discussions early in the project with the MAFF Project Officer, a sampling strategy was agreed and implemented. The intention of the sampling strategy was to establish how common hot particles are in the Sellafield area and to determine whether or not they are associated with foodstuffs for human or animal consumption. From this information, and a knowledge of the range of associated radionuclide activities, an assessment can be made of the radiobiological implications of these particles.

3. Sampling and Analysis

3.1 Confirming the presence of a hot particle

The presence of hot particles in terrestrial samples can be confirmed through the use of solid-state or emulsion track detectors. A sample is processed to obtain a thin, flat matrix which is presented to the appropriate detector medium. After a suitable exposure period, the detector is processed to reveal the nuclear tracks created, and these tracks are examined under a microscope. The presence of a hot particle is made apparent by the clustering of a large number of particle tracks in a small area, relative to the low number of tracks in its immediate surroundings. The activity of the hot particle can be estimated by considering the exposure time, the detector efficiency and the number of tracks observed. It is important to bear in mind that the track density in a cluster is governed as much by the exposure period as by the intrinsic activity of a hot particle. This time of exposure, whether to the track detector or to body tissue once a hot particle has entered an organism, is a major consideration in assessing the effects of hot particles.

3.2 Sample selection and collection

In order to quantify the abundance of radioactive particles in the terrestrial environment in the vicinity of Sellafield, a representative suite of grass and soil samples was collected in February and March of 1995. Studies of the fate of hot particles emitted from Chernobyl have shown that particles deposited on land that has since been subjected to cultivation are likely to have been rapidly destroyed (Konoplev & Bobovnikova 1991), or transported down the soil profile (Petrayev *et al.* 1992). After consultation with local farmers, sample sites for soil and grass were chosen. These were restricted to pasture land that had lain undisturbed for at least 30 years. This strategy was adopted to maximise the efficiency of the environmental survey, in terms of obtaining samples representative of the historical deposition of particles from Sellafield.

At each location sampled, five surface scrapes of 0-2 cm depth were taken from the centre and corners of a 1m square, producing five turves with a total area of 0.4 m^2. Where possible, the underlying soil layer (2-3 cm was also sampled, see Table 1). The turves were returned to the laboratory, where the grass was trimmed to within 1 cm of the soil surface, collected and oven dried at 35°C. The soil was oven dried at 35°C, gently disaggregated in a pestle and mortar, and

sieved to 2 mm to remove stones and vegetation. Dry weight yields were recorded to allow subsequent calculation of deposition per unit area.

Samples were also taken from three sites in Ladywood, a small coniferous wood to the north-east of Sellafield. The ground litter of this woodland had been identified as containing radioactive particles in a previous study (Day *et al.* 1992). A similar sampling protocol to that employed for soil and grass was adopted, with scrapes obtained from 0-2 cm and 2-5 cm. The samples were oven dried at 35°C and sieved to 0.5 mm, producing two fractions from each depth: 'needles' and 'fines'.

Efforts were then directed toward assessing the availability of radioactive particles to the food chain. Current arable land-use in the Sellafield area is quite restricted. Potatoes are the only crop grown commercially for human consumption. Wheat and barley are grown, but these crops are used solely for animal fodder. Most of the local farmland is given over to pasture for sheep and cattle. Though this might suggest that there is little risk of any radioactive particles being consumed directly by humans, a habit survey of the local population (Stewart *et al.* 1990) found that, for more than half the sample group, the majority of vegetables consumed were home grown.

The consumption of home grown vegetables may be potentially an important route into the human food chain for radioactive particles. It was therefore decided to establish vegetable plots close to the perimeter of Sellafield, rather than to obtain samples of locally grown produce directly from residents of nearby villages. In this manner, greater control could be exerted over growing conditions, ensuring consistency of treatment between the plots. Three such plots were prepared at secure sites in two cottage gardens and at the BNFL meteorological mast compound. A variety of leafy green vegetables, which had been seeded and grown in a nursery at Westlakes for a month, were planted out in early June. These consisted of ten plants each of broccoli, cabbage, spinach, lettuce and leek at each plot. It was decided to concentrate on green vegetables, rather than root vegetables, as leafy vegetables are considered more likely to intercept and retain freshly deposited or resuspended particles, whereas the preparation of root vegetables (scrubbing, scraping, peeling) is likely to minimise the risk of transfer of hot particles to consumers.

Vegetables produced from two of the three vegetable plots established close to Sellafield were successfully harvested in late September, with the exception of the broccoli which, due to the unusually hot and dry summer, bolted before remedial action could be taken. The entire crop on the third plot was lost after a particularly severe caterpillar infestation.

With a view to differentiating between freshly and historically deposited particles, two grass trays and a 'frisbee' - a large surface area deposition collector (1 metre diameter) - were deployed on the site of each vegetable plot. Samples were collected from the frisbees and one of the grass trays at each site on a monthly basis over a four month period. The grass from the other grass trays was harvested at the end of the four months.

This suite of samples was expanded by the addition of blackberries collected from the vicinity of Calder Hall, and samples of barley and wheat from Seascale Farm. It was also intended that the stomach from a cow, which had died on a local farm, be examined for evidence of the presence of radioactive particles. Unfortunately, the stomach, which was in cold storage at the Central Veterinary Laboratory, had been mislaid and was thus unavailable for analysis.

The samples that were collected are described in Table 1 and the locations of the sample sites are shown in Figure 1.

3.3 Sample processing

Sub-samples of the soil and grass, collected in the initial environmental survey and the monthly sampling of the grass trays, were presented to sections of Kodak LR-115® cellulose nitrate α-radiation sensitive film (6 cm × 9 cm), in the form of dried, homogenised material, spread thinly upon adhesive mounting card. Good contact between sample and film was ensured by sandwiching the cards between perspex plates in a press for periods of 4, 8 and 12 weeks. The performance of this plastic detector has been demonstrated in several studies prior to this one (e.g. Qaqish and Besant, 1976; Baxter *et al.* 1989).

The Ladywood litter samples were processed in a similar manner to the soils and grass.

Table 1. Sampling locations and sample inventory

	Locations of sample sites	OS grid reference	Nature of samples
1	Church House Farm A	3035 5045	Grass, Soil 0-2 cm, 2-3 cm
2	Church House Farm B	3041 5043	Soil 0-2 cm, (grass too short to cut)
3	Mid Tarn Farm	3023 5042	Grass, Soil 0-2 cm, 2-3 cm
4	Calder Farm A	3038 5037	Grass, Soil 0-2 cm
5	Seascale Farm A	3039 5029	Grass, Soil 0-2 cm
6	Seascale Farm B	3038 5029	Grass, Soil 0-2 cm
7	Greenmoorside Farm	3022 5054	Grass, Soil 0-2 cm
8	Tarn Head Farm	3023 5054	Grass, Soil 0-2 cm
9	Ladywood A	3036 5033	Pine needle litter 0-2 cm, 2-5 cm
10	Ladywood B	3037 5035	Pine needle litter 0-2 cm, 2-5 cm
11	Ladywood C	3037 5037	Pine needle litter 0-2 cm, 2-5 cm
12	BNFL Meteorological Mast	3022 5045	Grass, Soil 0-2 cm, grass trays, frisbees, 6 cabbage, 6 lettuce, 7 leek, 7 spinach.
13	Yottenfews Cottage Garden	3032 5051	Grass, Soil 0-2 cm, grass trays, frisbees, vegetable plot destroyed by caterpillars.
14	Stang's Cottage Garden	3038 5035	Grass, Soil 0-2 cm, grass trays, frisbees, 7 cabbage, 5 lettuce, 8 leek, 8 spinach.
15	Seascale Farm C	3042 5031	Wheat
16	Seascale Farm D	3041 5025	Barley
17	Calder Hall	3035 5038	Blackberries

The vegetable samples were treated slightly differently. Each vegetable was sliced or broken up as if in preparation for consumption and then the portions thoroughly washed in 3 litres of water, with the aim of removing particulate material adhering to the plant surfaces. This wash water was filtered (Whatman GF/C), the filters dried, and LR-115® film exposed to the dried filter paper for a period of 4 weeks. The vegetable material was then dried, homogenised and presented to LR-115® film in the same manner as the soil and grass samples, though instead of sub-samples being exposed for 4, 8 and 12 weeks, three sub-samples were exposed for 4 weeks.

The grain samples were simply washed, and the washings filtered to obtain filter paper samples, whereas the blackberries were dried and homogenised to give card samples.

The frisbee boards were rinsed thoroughly, and the rinse-water filtered to yield filters that were processed in the same manner as the vegetable rinse water.

In this manner, every sample, apart from the washings of the vegetables and the frisbee boards, was analysed in triplicate. Table 3 provides a summary of the sample exposures that were made.

Table 3. Summary of sample exposures

Sample Type	Number	Sample Type	Number
Soils	39	Vegetable	162
Grass (from pasture)	30	Vegetable washings	54
Grass (monthly trays)	27	Wheat	3
Grass (4-month trays)	9	Barley	3
Pine litter (needles)	18	Blackberries	9
Pine litter (fines)	18	Frisbee washings	12
Total Number of Exposures = 381			

3.4 LR-115® analysis

After the required period, the LR-115® plastic from each subsample was developed by etching in 2.5M sodium hydroxide at 60°C, for approximately 90 minutes. This process preferentially enlarges the sub-microscopic damage tracks produced as incident alpha-particles penetrate the plastic. Each plastic film was then examined under an optical microscope at ×20 magnification. The presence of a radioactive particle within the sub-sample matrix was made apparent from the dense clustering of tracks produced by the alpha-radiation emitted from the particle.

4. Results

4.1 Results of LR-115® Analysis

No examples of the dense clustering of damage tracks that would indicate the presence of radioactive particles were observed in any of the films exposed to samples from soils, leaf litter, vegetables, grains or blackberries. This was also true of the films exposed to the washings from the frisbees and the grass obtained from the grass trays.

Evidence of two particles generating alpha-track densities greater than background was discovered in fresh grass samples, one taken from Mid Tarn Farm (site 3) the other from Seascale Farm B (site 5). Examination of the film from the four-week exposures of the samples revealed the presence of 'burned-out' regions surrounded by a dense halo of tracks, approximately 50 - 70 μm across in a pattern suggestive of damage due to alpha-emissions from a small, nearly spherical radioactive particle. Re-exposure of the samples for one week revealed similar patterns in the same locations, confirming the presence of a highly localised zone of alpha-activity within the matrix of each sample. Photographs of these exposures were taken and studied (Figs 2a and 2b). Photographs were also obtained from 72 hour exposures of the samples, which permitted the "burned-out" areas observed in Fig 2 to be resolved into individual tracks (Figs 3a and 3b).

4.2 Scanning electron microscopy and electron probe microanalysis

Five-millimetre square sections, corresponding to the burn-out zones in the LR-115® film, were cut from the two sample cards for examination under a Cambridge Instruments 360 scanning electron microscope. Initially, each sample section was viewed at low-magnification using back-scattered imaging (BSI). In this mode, judicious balancing of brightness and contrast can highlight regions containing concentrations of heavy elements (Sandalls & Baker, 1995).

When viewed under BSI, several hundred bright spots were apparent on both of the sample sections. A systematic survey was undertaken in which of each of the objects responsible for these bright spots was subjected to electron probe microanalysis (EPMA), to obtain information

on their elemental composition. This revealed that the vast majority of these were found to be iron-bearing soil particles. A typical energy dispersive x-ray spectrum is shown in Fig 4. Such particles are unlikely to have caused the alpha-tracks observed in the LR-115® exposures. Two particles of differing composition were, however, located in the two sample sections, and these are discussed in greater detail below.

4.2.1 Examination of particle from Mid Tarn Farm: particle no. 1

A secondary electron image of particle 1 is shown in Fig 5a. The image below was obtained using BSI (Fig 5b). The particle is approximately 20 μm long by 5 μm wide, with a very smooth surface.

The energy dispersive spectrum of particle 1 is shown in two parts in Figs 6a and 6b. Figure 6a shows the magnified spectrum in its entirety, in which a peak corresponding to thorium is apparent. Figure 6b gives an expanded view of the low-energy half of the spectrum, revealing very strong signals for phosphorus, lanthanum and cerium. It is probable that the overlapping peaks of lanthanum and cerium conceal lesser peaks from other lanthanides.

The spectrum is unique among all those examined, and the elemental composition suggests that the particle is of natural origin. Thorium, an alpha-emitting nuclide, is often found associated with the lanthanides, in minerals such as monazite, a phosphate of thorium, lanthanum and cerium. The monazite structure consists of distorted PO_4 tetrahedra with each metal atom roughly equidistant from nine oxygen atoms (Deer *et al.* 1992). This particle may be of local mineral origin; relatively high concentrations of lanthanum, mostly as monazite, are present in stream sediments close to Sellafield (British Geological Survey, 1992). Another explanation for the origin of the particle is the use of fertilisers in the study area. Certain fertilisers can be traced back to the original phosphate ore used in the manufacturing process and hence the characteristic radionuclide/elemental make-up of it could be derived. However, since details of fertiliser use in the area are not forthcoming, it is difficult to pursue this line of explanation further. One further point that has been considered is that there is a phosphoric acid plant at Whitehaven that used to process phosphate ore up to the early 1990's (Poole *et al.* 1995). Since the distance between this plant and the sample area is more than 10 km it is unlikely that particles of the size discussed

here could have travelled that far. It seems reasonable to deduce that this particle, probably of natural origin, is responsible for the cluster of alpha-tracks observed in the LR-115® exposure of this sample.

4.2.2 Examination of particle from Seascale Farm B: Particle no. 2

A secondary electron image of particle 2 is shown in Fig 7a and its back-scattered image is shown in Fig 7b. Particle 2 is of a similar size to particle 1, but is of entirely different composition. Its energy dispersive x-ray spectrum is shown in Figure 8, and from this it appears that the particle is composed mainly of zirconium.

This particle is a possible candidate for the alpha-emitter detected by the LR-115®. The strong zirconium signal suggests the possibility that the particle may be a fragment of fuel cladding, but the lack of a peak for uranium or plutonium in the energy dispersive x-ray spectrum means that no firm conclusion can be made. It must be emphasised that EPMA provides information only on the surface of the object being studied, so the alpha-emissions could originate from a region beneath a coating of zirconium. The absence of a signal for silicon rules out the possibility that the particle could be a mineral fragment, such as zircon.

4.3 Results of alpha-spectrometry

The spectra of the isolated sample sections did exhibit alpha-activity above background levels, but the spectra were too degraded to obtain any useful qualitative or quantitative data.

5. Discussion

With some assumptions, an estimate of the activities of the two particles can be made. In experimental work on Chernobyl hot particles, Kushin *et al.* (1993) determined a registration efficiency for LR-115® of $\rho = 0.47$, assuming that the particles being studied were spherical and on the surface of the sample. The electron micrographs show that, in both samples, the particles are very close to the sample surface. Some assumptions about the geometric efficiency (γ) must

also be made. Examining the immediate environment of each particle, it can be seen that both are in close proximity to other, larger, particles. These will certainly occlude some proportion of the alpha-radiation being emitted from the active particles. A figure of $\gamma = 0.35$ would seem to be a reasonable estimate for the geometric efficiency. The activities of the particles can then be estimated using the following equation:

$$A = \frac{x}{\rho.\gamma.t}$$

where; A = activity of the particle (Bq),

x = number of tracks counted,

ρ = registration efficiency,

γ = geometric efficiency,

t = exposure time (s).

The photographs of the 72 hour exposures revealed approximately 55 tracks for Particle 1 and 45 tracks for Particle 2. Putting these figures into the above equation gives total activities of 1.3 mBq for Particle 1 and 1.1 mBq for Particle 2. It must be emphasised that this is a crude calculation, but it may give a first order approximation of the activities of the particles in the absence of useful spectrometric data.

Only two radioactive particles, of low activity, were found in almost 400 exposures of samples, suggesting that abundance in the immediate vicinity of Sellafield is very low. Both were found on grass samples from undisturbed pasture, not on any foodstuffs for direct consumption by humans. The thorium-bearing particle appears to be a fragment of local mineral origin, probably monazite. If the zirconium-bearing particle is responsible for the alpha-tracks observed on the film then it is certainly of Sellafield origin. It is not possible, however, to determine whether the particle had been freshly deposited upon the grass, or whether it had its origin in some historical discharge.

Such particles may be available for ingestion by cattle or sheep, but because the behaviour of radioactive particles *in vitro* is not at all well understood, it is difficult to assess the extent of any hazard to humans consuming meat or offal from an animal exposed in this manner. The size of the particles precludes the possibility of inhalation and lodging in the deep lung, either by

humans or animals, as the upper size limit for the deposition in the deep lung of inhaled particles is approximately 10 μm (ICRP 1994). Such a particle could be inhaled into the nasopharyngeal tract but would then be fairly rapidly removed with the mucosal secretions.

Since no hot particles were associated with any of the food items investigated here then, strictly speaking, the radiological impact of hot particles is insignificant in terrestrial foodstuffs cultivated in the vicinity of the Sellafield plant. Hypothetical calculations, however, have been performed in order to provide some indication of the doses that could be associated with the radioactive particles encountered in this study.

Calculations were performed using LUDEP (Jarvis & Birchall 1994) which incorporates details of the lung model described in ICRP 66 (ICRP 1994) and the metabolic models included in ICRP 30 (ICRP 1979). Since the activities of the particles were of the order of 1 mBq, the particles have been treated as typically inhaled or ingested particles. Calculations were based on the particle possessing 1 mBq of either ^{232}Th or ^{238}U, the particle being either inhaled or ingested, classed as having either a medium or slow residence time in the body and that the final dose refers to the committed dose over the period of 50 years to an adult outdoors. No calculations were performed for external doses since no information on the beta/gamma activities of the particles was available. The various calculations showed similar values for the Th and U scenarios and are summarised in the table below.

Table 3. Summary of dose calculations

Exposure scenario	Committed dose (over 50 years)
Maximum dose to respiratory tract - nasal passage	8 nanoSieverts
Maximum organ dose - bone surface	1 milliSieverts

The insignificance of these calculated doses is emphasised when it is borne in mind that the average annual dose in the UK from all natural sources is 2.2 mSv (Hughes & O'Riordan 1993).

6. Conclusions

- The abundance of radioactive particles in the vicinity of Sellafield is low. More than 100 environmental samples were collected, over a range of 10 to 1000 metres from the site perimeter. Evidence was found for only two active particles in almost 400 autoradiographic exposures.
- Estimates for the activities of the two particles are of the order of 1 mBq. Of these two, one appears to be a fragment of a naturally occurring thorium-bearing mineral, probably local monazite. The other was not positively identified, but a possible explanation is offered by the presence of a zirconium-rich particle, which may be associated with nuclear fuel. Both particles are of a similar size, approximately 20 µm long by 5 µm wide.
- No evidence for any radioactive particles was found on foodstuffs available for consumption by humans. The two active particles were discovered on samples of grass from pasture, and so could be ingested by livestock.
- Even if inhaled or ingested, the hypothetical maximum dose delivered by such a particle is insignificant when compared with the doses from natural radiation. Calculations yield a conservative upper limit for the committed dose from an inhaled particle of 1 mSv, over 50 years, to the bone surface.

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Fig 1. Map of Sellafield area showing sample locations

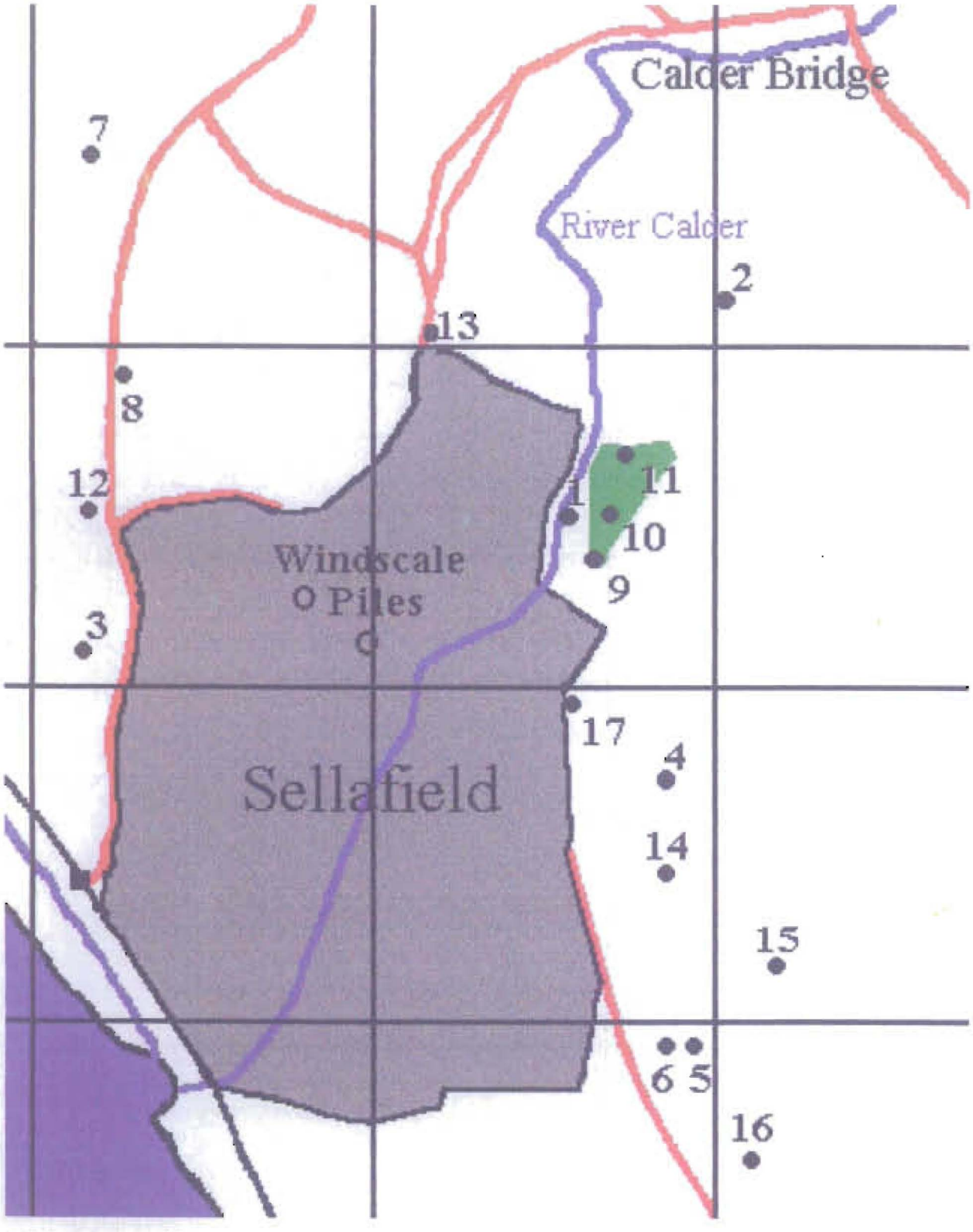


Fig 2a. Autoradiograph of active particle from Mid Tarn Farm (4 week exposure)

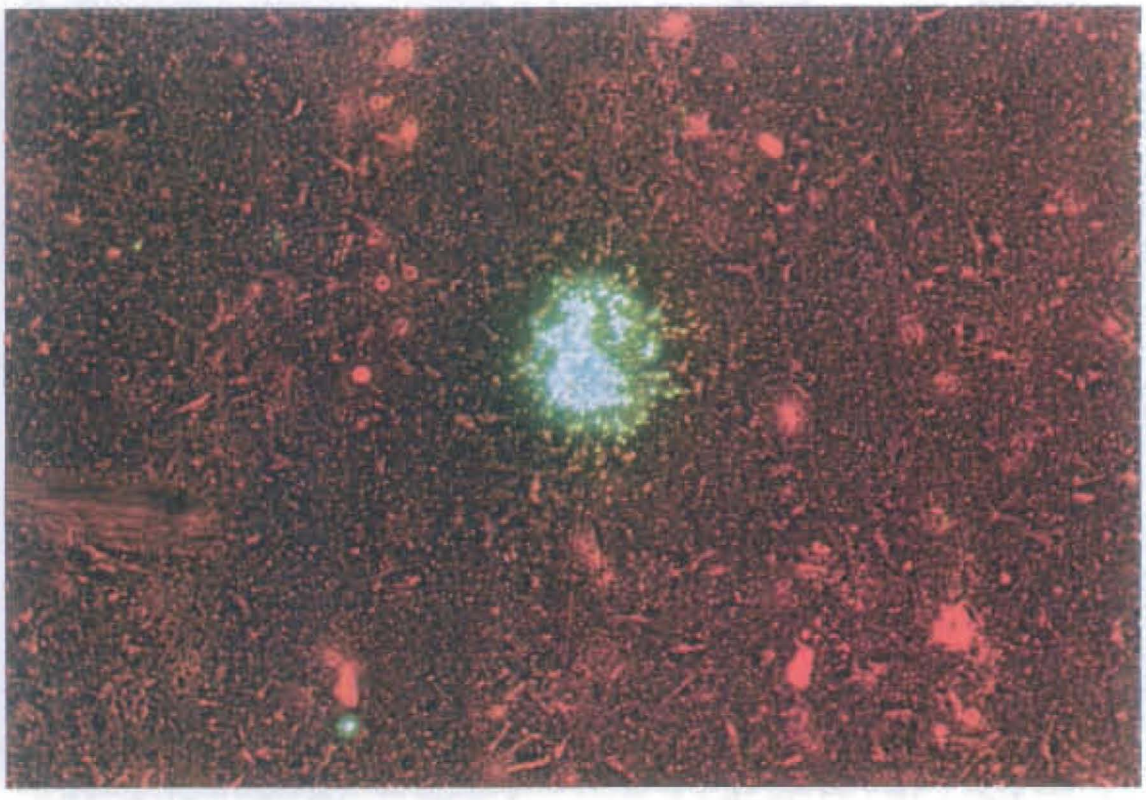


Fig 2b. Autoradiograph of active particle from Seascale Farm B (4 week exposure)

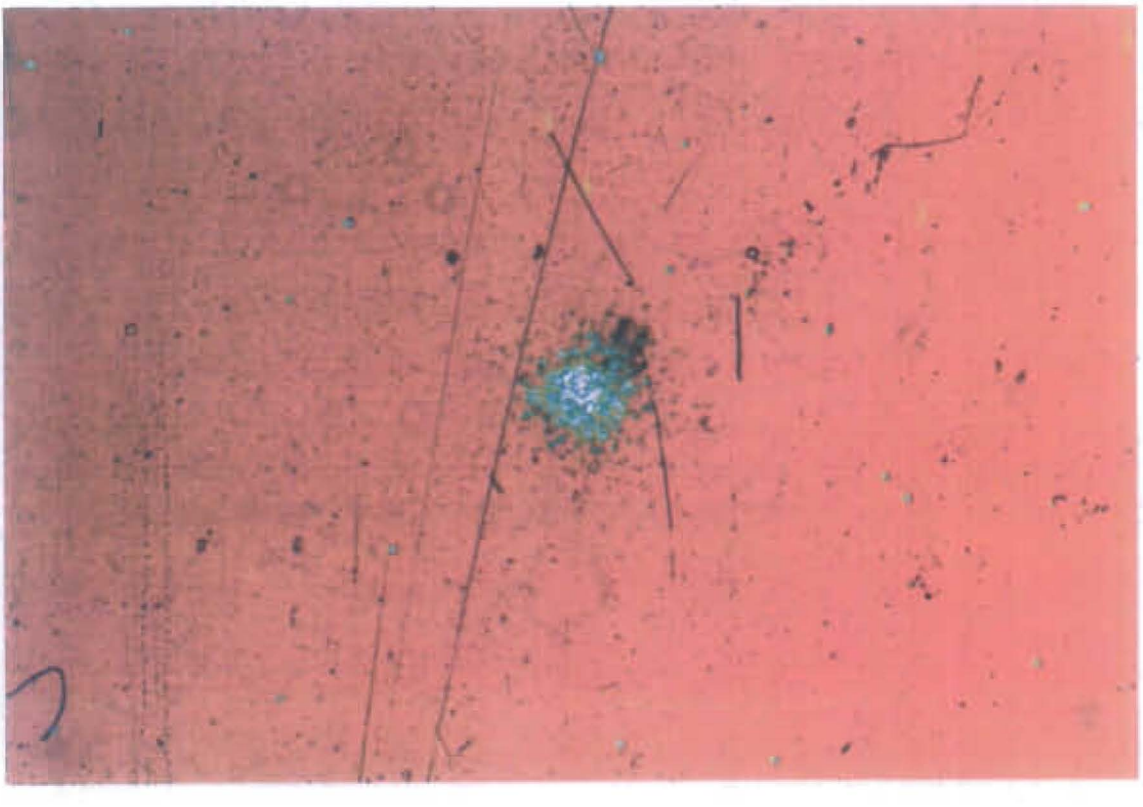


Fig 3a. Autoradiograph of active particle from Mid Tarn Farm (72 hour exposure)

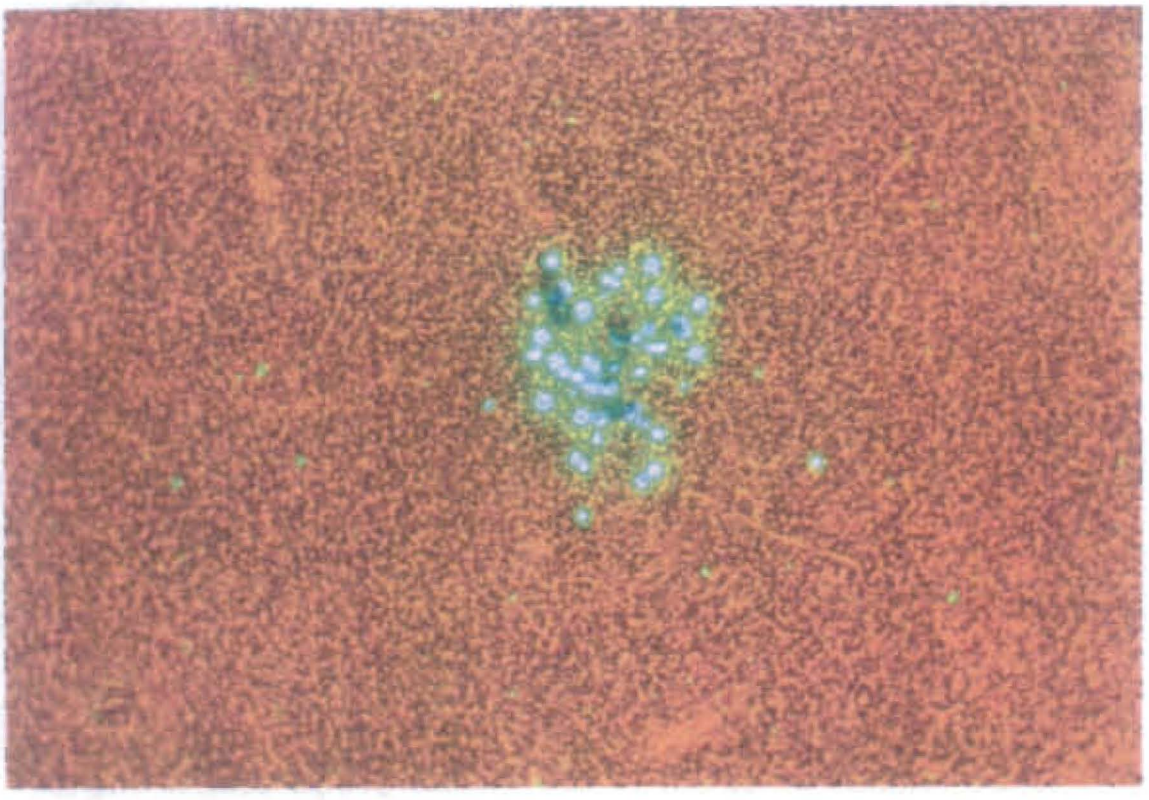


Fig 3b. Autoradiograph of active particle from Seascale Farm B (72 hour exposure)

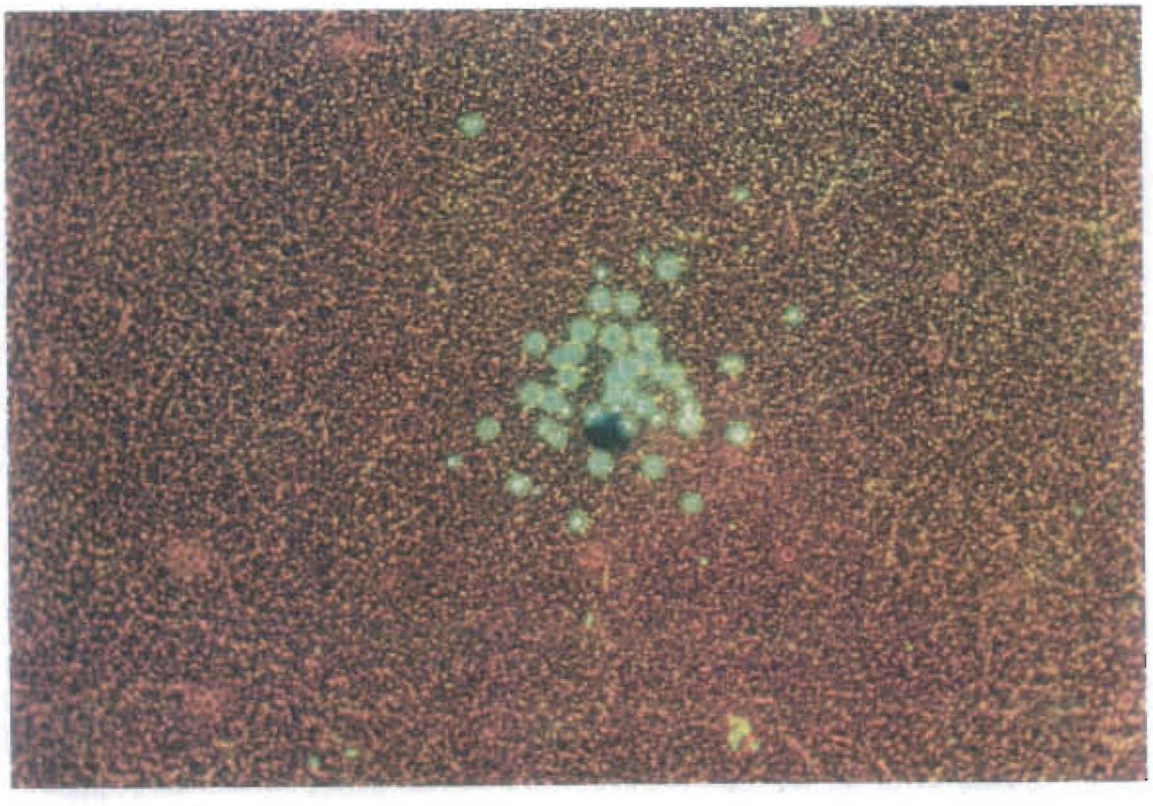


Fig 4. Energy dispersive x-ray spectrum of iron-bearing particle

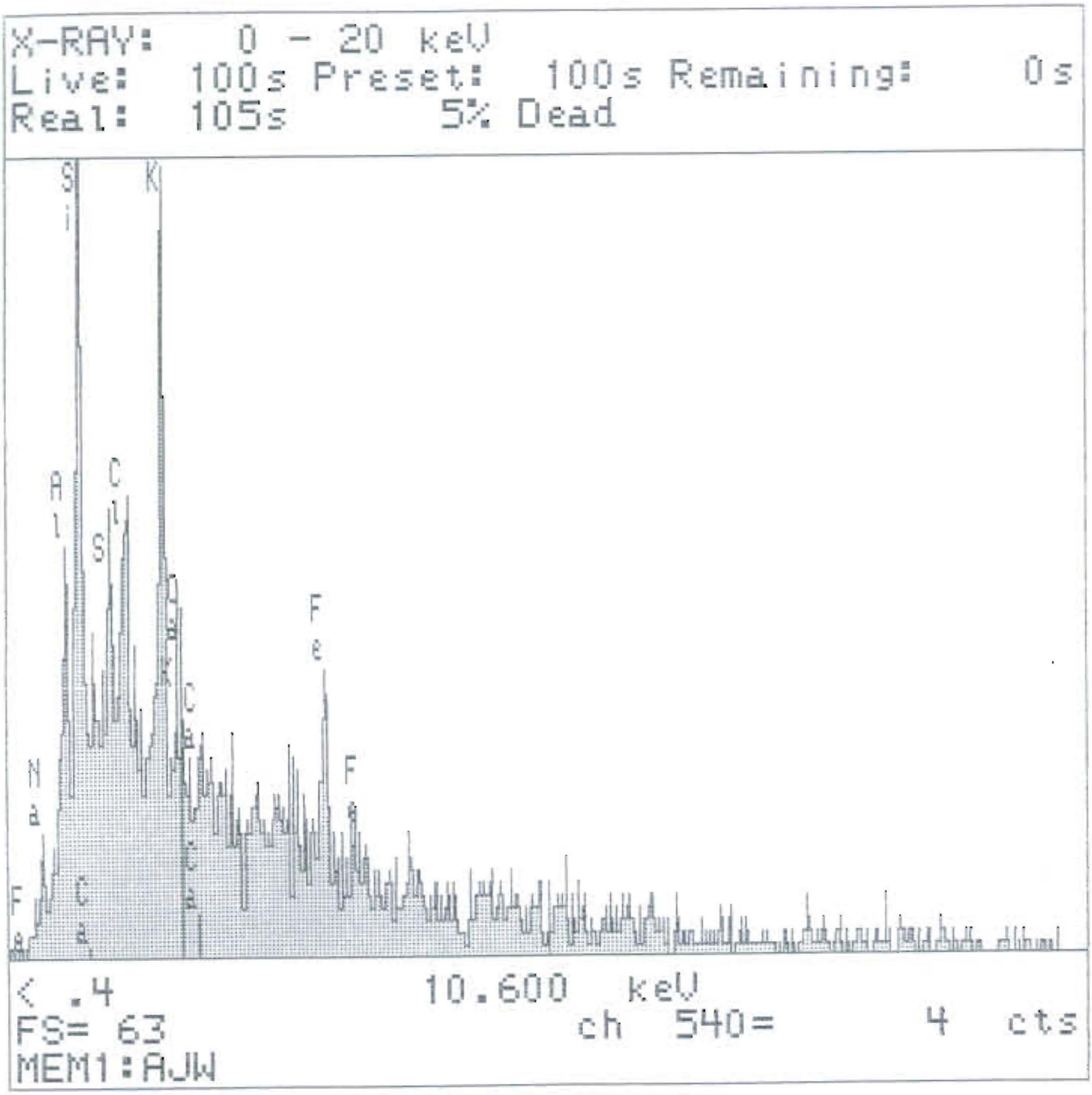


Fig 5a.Secondary electron image of Particle No.1

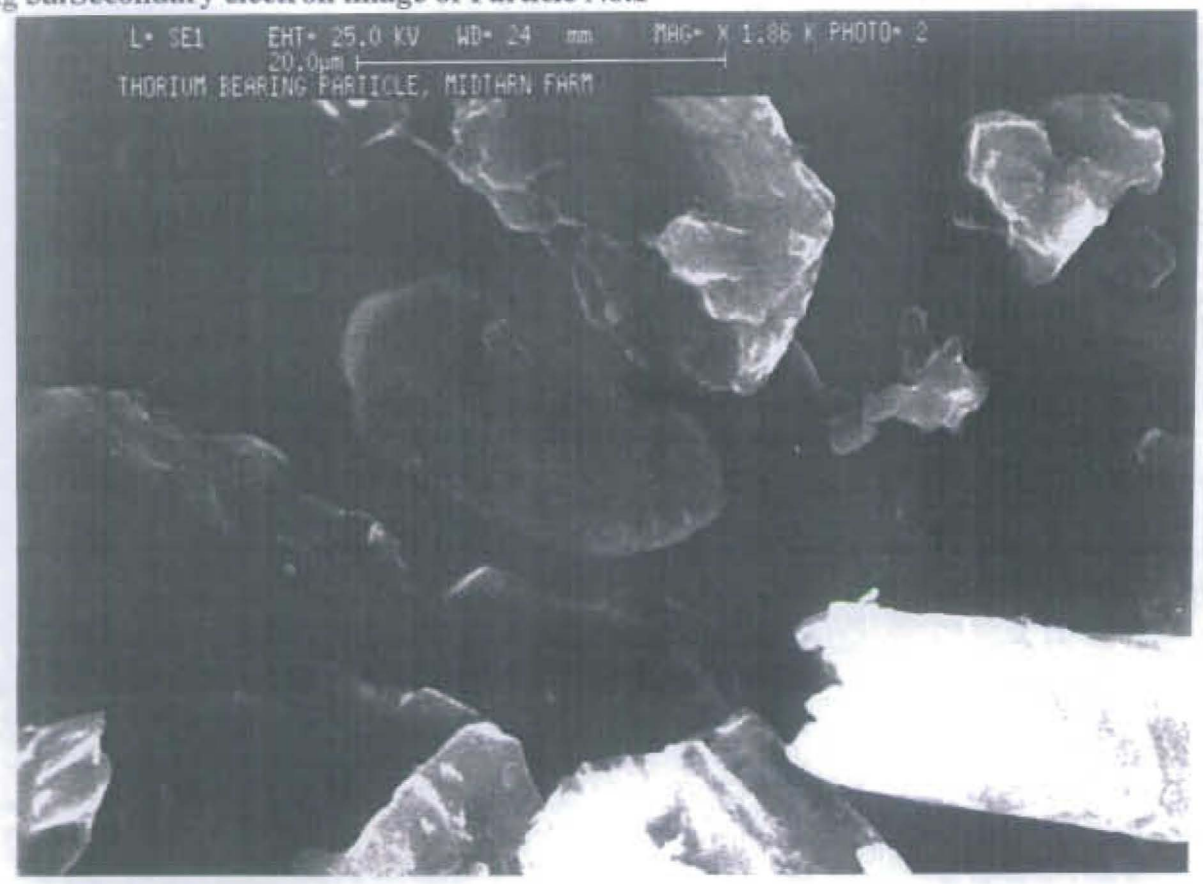


Fig 5b.Back-scattered electron image of Particle No.1

Fig 6a. Energy dispersive x-ray spectrum of Particle No.1

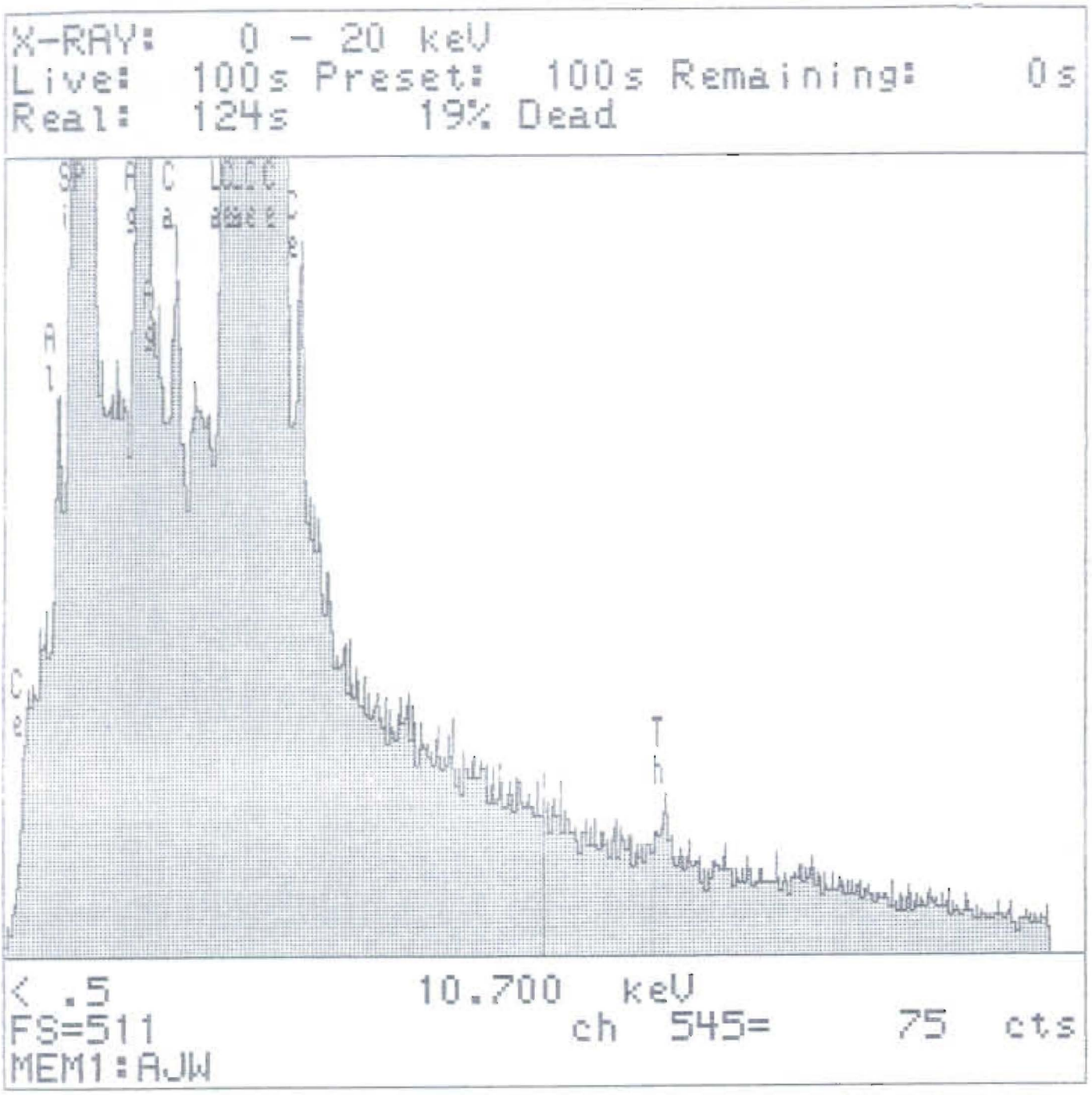


Fig 6b. Energy dispersive x-ray spectrum of Particle No.1

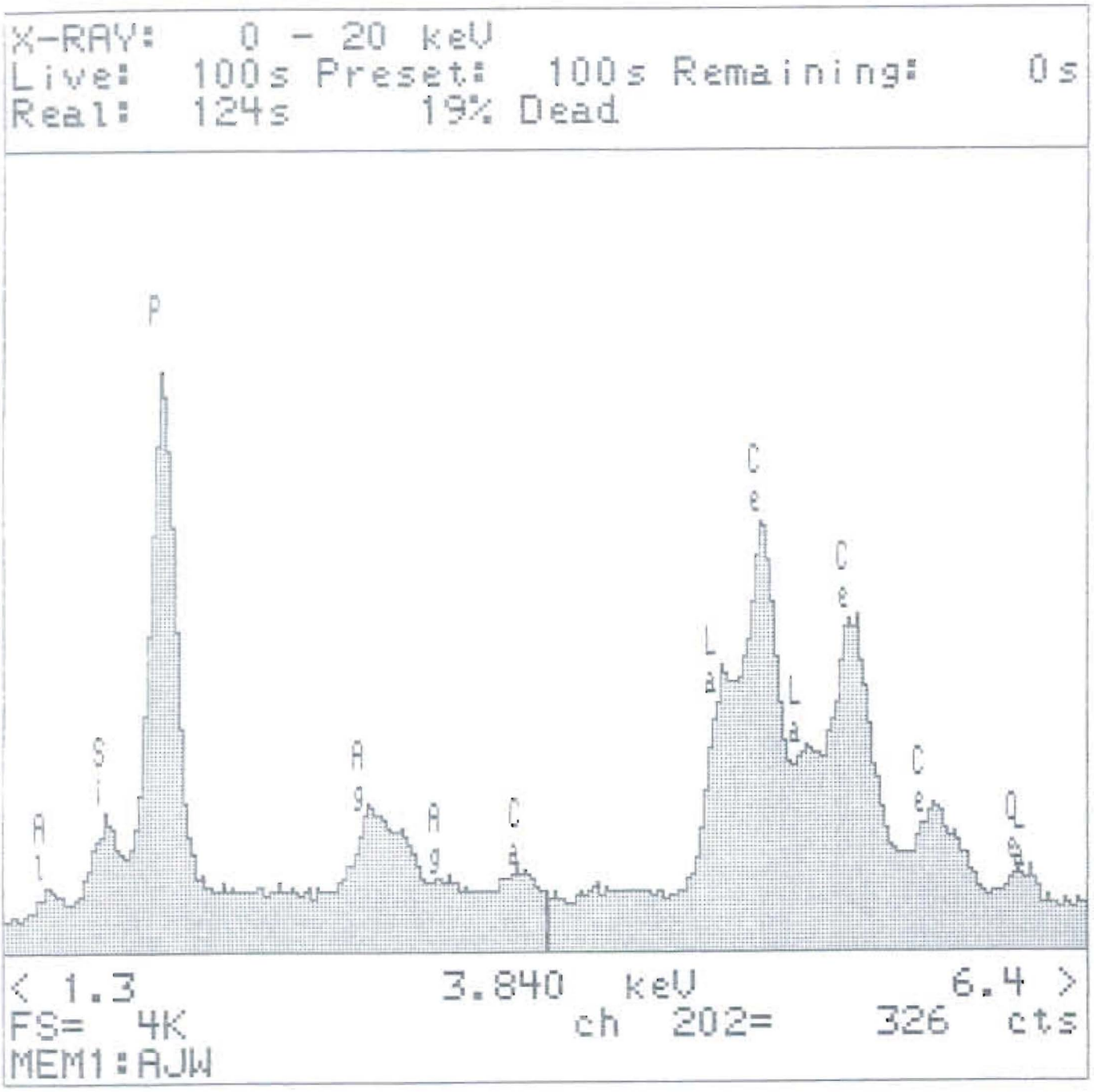


Fig 7a.Secondary electron image of Particle No.2

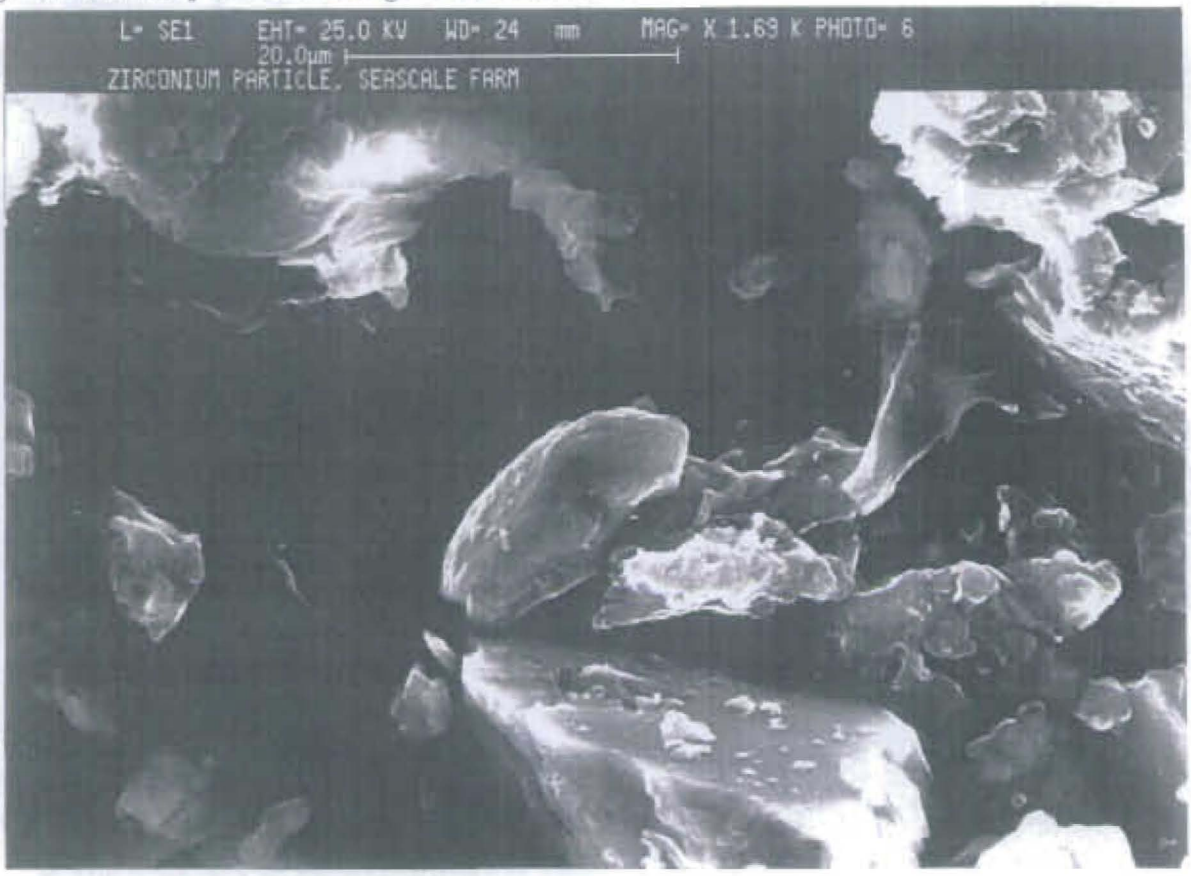


Fig 7b.Back-scattered electron image of Particle No.2

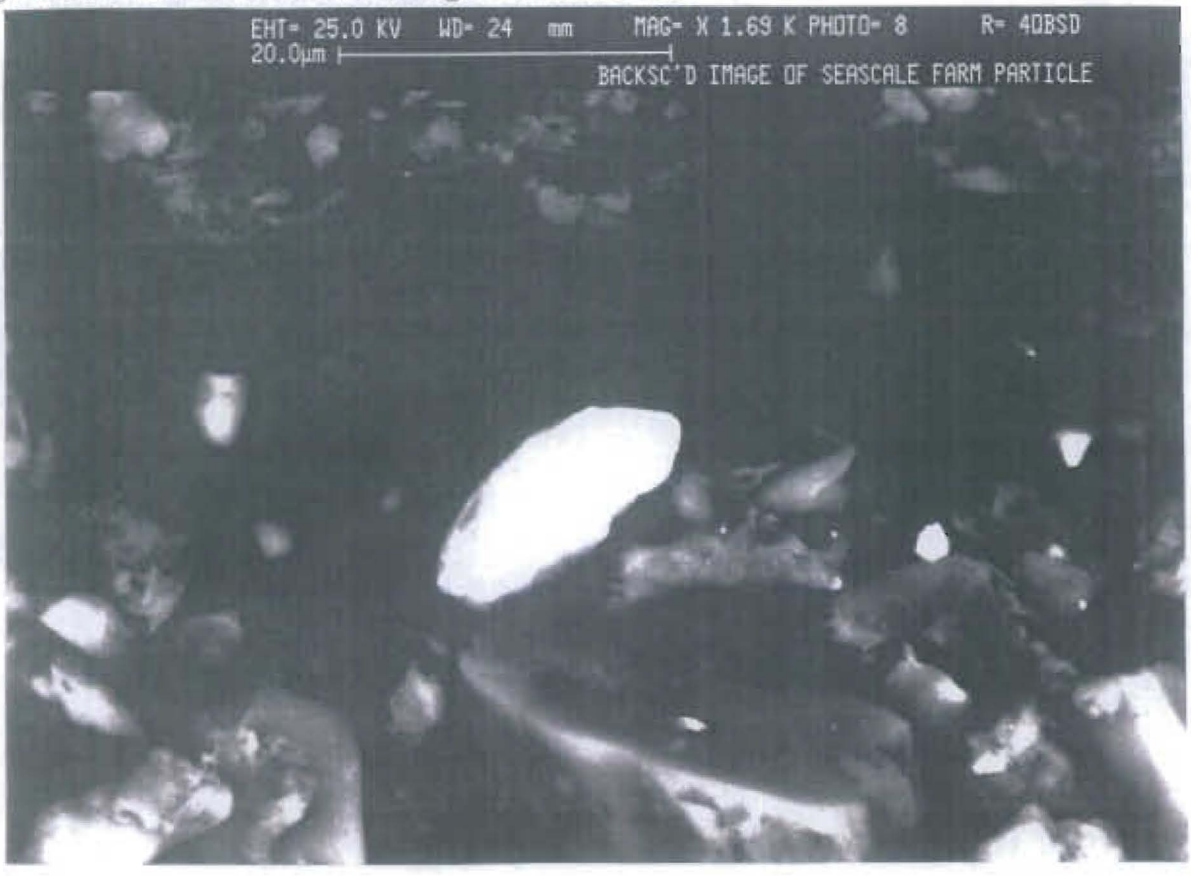
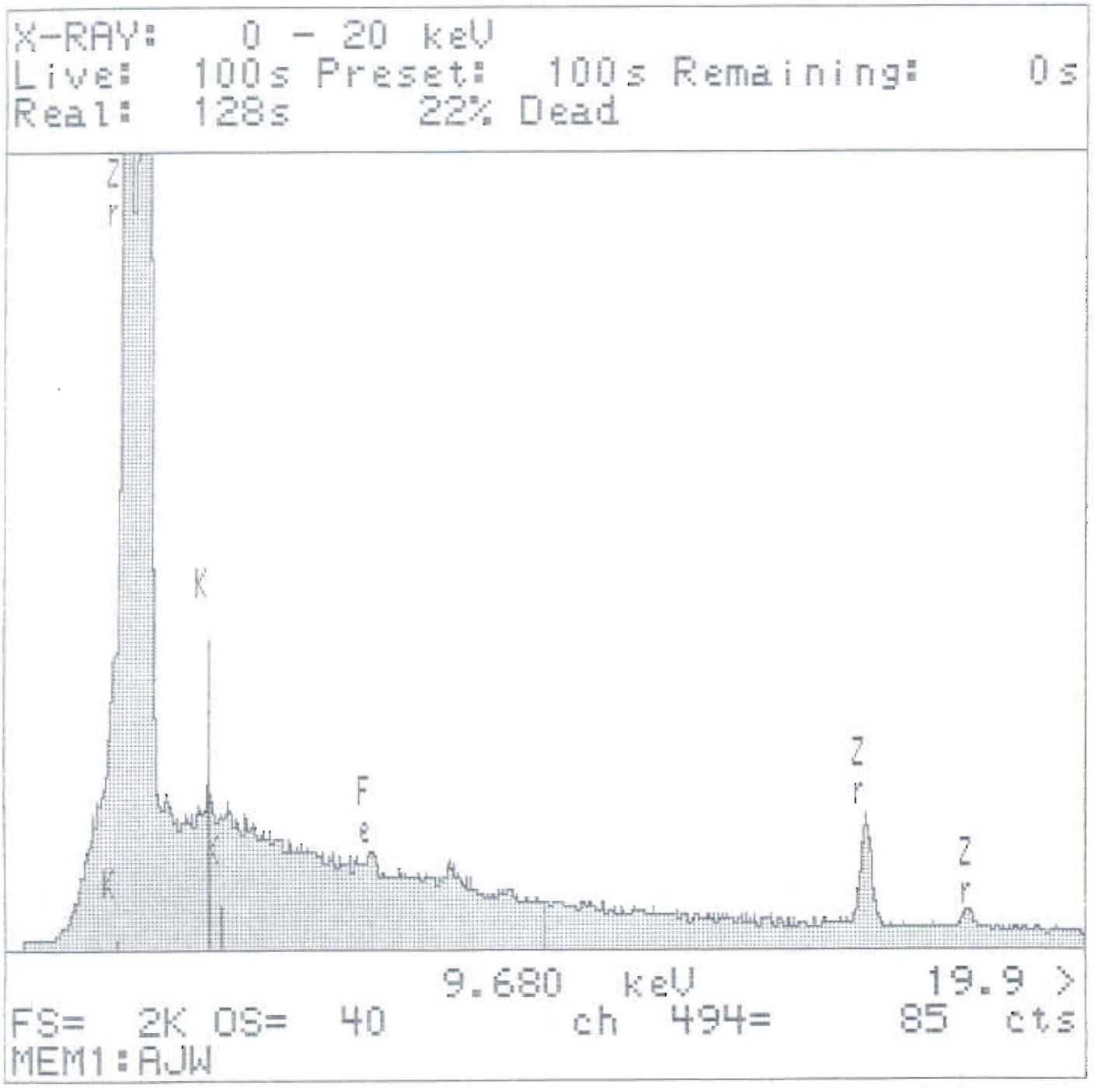


Fig 8. Energy dispersive x-ray spectrum of Particle No.2



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