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Journal of Nuclear Materials 324 (2004) 1–5

journal of nuclear materials

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Production of uranium–molybdenum particles by spark-erosion

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Received 5 May 2003; accepted 20 August 2003

Abstract

With the spark-erosion method we have produced spheroidal particles of an uranium–molybdenum alloy using pure water as dielectric. The particles were characterized by optical metallography, scanning electron microscopy, energy dispersive spectrometry and X-ray diffraction. Mostly spherical particles of UO_2 with a distinctive size distribution with peaks centered at 70 and 10 μm were obtained. The particles have central inclusions of U and Mo compounds.

1. Introduction

The spark-erosion or electrodischarge machining process (EDM) is a worthwhile technique [1,2], used to cut and conform conductive materials by means of electric discharges [3]. It is mainly a thermal process which involves several facets like heating, melting, vaporizing, diffusion and fast cooling, all of them far from equilibrium. The operation consists of a swift energy strike on a reduced area, led by a pulsed power source. The energy surface density is very high and the temperature on the surfaces rapidly reaches the melting point and also the boiling point of the electrodes. A bubble is formed on the electrodesá surface and when it explodes very small spherical droplets are ejected from the condensed vapor of the bubble [4], and owing to the created depression, part of the liquid metal remaining on the surface is dispersed into the dielectric. It is suddenly solidified acquiring in general the spherical shape of the larger particles [5], acquiring a bimodal particle size distribution, [6]. Craters appear on the surfaces of the electrodes forming a resolidified layer known as white zone, similar to that formed by welding. The material in the layer is very different from the rest of the electrodes not only by the solidification process but for the incorporation of elements belonging to the dielectric as well. Crater size, composition, and thickness of the white zone depends on the current, duration of the discharge and dielectric [7–9].

The ejected material of the EDM is the main object of this work and based in our experience of producing spherical particles of iron [6], the feasibility of extending the method towards the production of uranium–molybdenum (U–Mo) particles is explored. In the present work, the first results that were obtained were aimed at preparation of spherical uranium alloy particles of interesting size for nuclear applications in a program aimed at conversion of high-enriched uranium (HEU) to low-enriched uranium (LEU) for use in a research reactor fuel.

2. Experimental

The electrodes were made of a depleted uranium lump of 99 wt% and molybdenum wires of 99.9 wt% induction-melted in a cylindrical graphite crucible

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doi:10.1016/j.jnucmat.2003.08.040

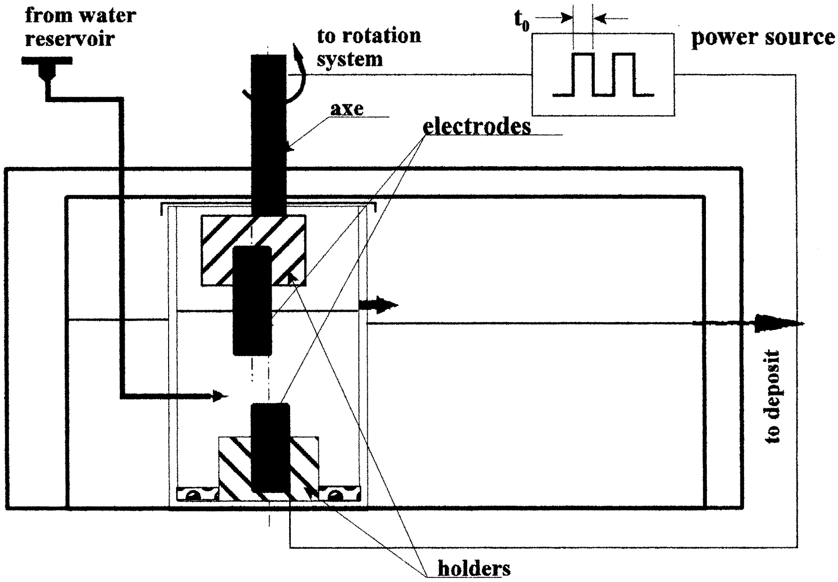

Fig. 1. Device showing the electrodes, holders, container boxes and water line.

previously coated with a heat resistant ceramic. A way a rod of 6 mm diameter and approximately 90 mm length was obtained which was cooled in air at room temperature and cut into two pieces of 50 and 40 mm length each. Our experience was performed employing a commercial electrodischarge machine (CT Electromecänica Ltd.®, Argentina) using a current of 25 Amp and an active pulse time, t_0, of 2048 μs, cf. Fig. 1. The dielectric was demineralized water prepared by inverse osmosis and had a resistivity of 1 MΩ. A scanning electron microscope (SEM), Philips 500, was employed to examine the powder morphology. The chemical composition was evaluated by energy dispersive spectroscopy (EDS). The powder density was measured by helium picnometry in a 'Quantachrome' equipment. The crystal structure of the powder was analyzed by X-ray diffraction using Cu-Kα radiation on the surface and the inner parts of the particles. For some analysis the particles were immersed in a epoxy resin and polished to observe its interior characteristics.

3. Results

The metallographic structure of the 'as melted' material alloy is displayed in Fig. 2. The shape of the grains are rounded, indicating a rather good recrystallization during cooling in air. The grains present some inclusions, due to the purity of the alloy components. The grains near the surface of the rod are smaller than those of the midpoint due to different cooling velocities. The electrodes average composition, obtained by EDS, was: U 90 wt% and Mo 10 wt%, ±3 wt%.

The X-ray diffraction analysis of the rod, cf. Fig. 3 confirms the formation of the corresponding phase γ of the U–Mo phase diagram.

The yield of the collected material was about 13.6 g in 8 h work. The picnometry measurement was 9.86 ± 0.05 g cm^{-3}. The material lost was about of 0.15 g in all the process.

The amounts of the electrodes consuming is indicated in Table 1.

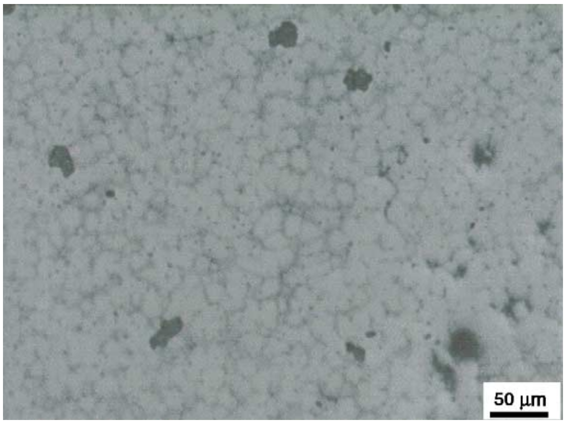

Fig. 2. Microstructure of the as melted electrodes, etched with one part of HNO_3, one of acetic acid and two of H_2O.

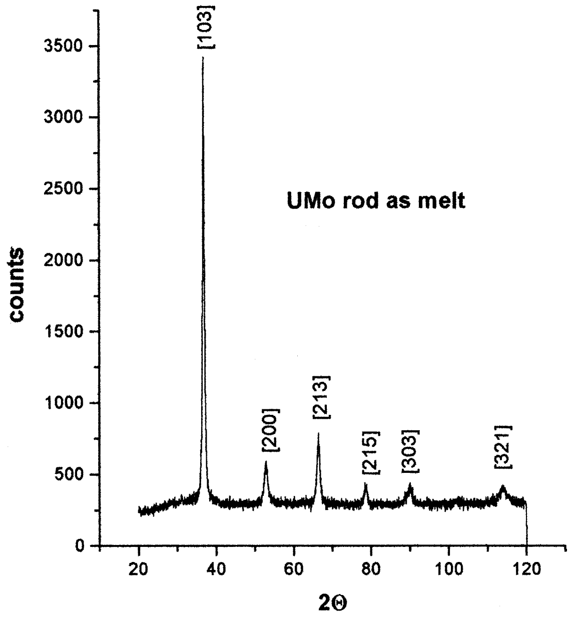

Fig. 3. Cu-Kα XRD of the as melted electrodes, lines correspond to U–Mo gamma phase.

Table 1
Electrodes consuming

Electrode	Initial (g)	Final (g)	ΔMass (g)	Effectiveness
Upper	21.68	11.43	−10.25	0.47
Lower	30.29	27.09	−3.20	0.11
Total	51.97	38.52	−13.45	0.26

By optical microscopy and by SEM we found that most of the particles have a relative regular smooth surface, cf. Figs. 4 and 5. The geometry of the particles is in general somewhat spherical. The nonspherical fraction appears to have been milled by the eccentric movement of the electrodes after freezing.

The size distribution of the particles shown in Fig. 6 is bimodal. The majority of the larger particles are about 70 μm and the smaller ones about 10 μm diameter.

Fig. 7 shows a particle with holes, the external layer of the particle shows a structure similar as those found in the oxidized iron debris of cutting laser, [10] material belonged cf. Fig. 8. In general the inner part of the spheres is not uniform, some of the spheres have large inclusions, Figs. 9 and 10. The EDS results of the internal part of the particles show that the nucleus – the dark zone cf. Figs. 9 and 10 of the particles do not exhibit a uniform composition, see Table 2.

The phases present, as indicated by X-ray diffraction, in the internal part of the particles are: strong presence of UO_2 with lesser amounts of pure U and U_2O_3; in the external part of particles: strong presence of UO_2, with lesser amounts of pure Mo and U, and even less amounts of MoU_2, cf. Fig. 11.

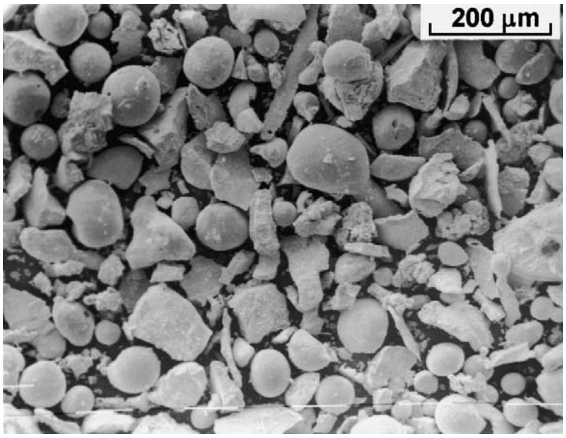

Fig. 4. SEM micrograph of particles made by electroerosion.

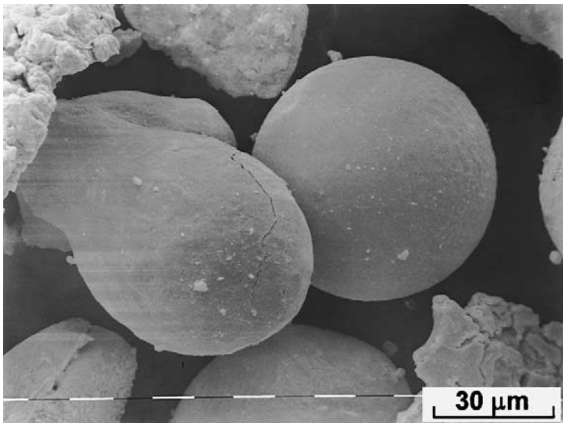

Fig. 5. SEM micrograph of spheroidal particles made by electroerosion.

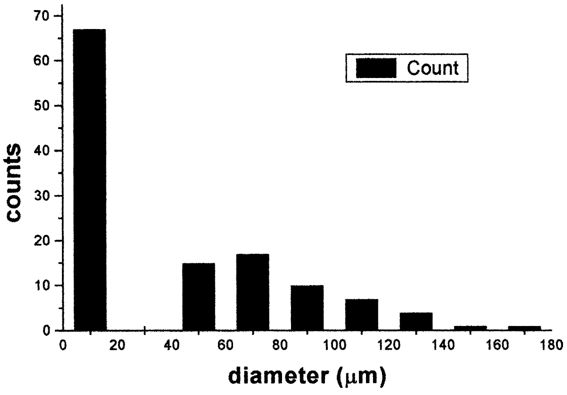

Fig. 6. Histogram of the electroeroded size of uranium–molybdenum particles.

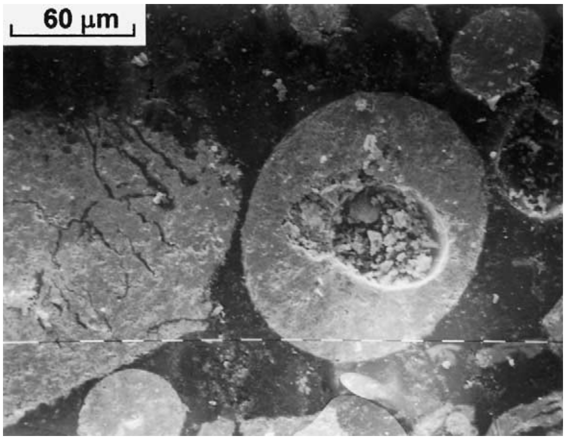

Fig. 7. Ball with a hole produced by an removed inclusion during the process of smoothing.

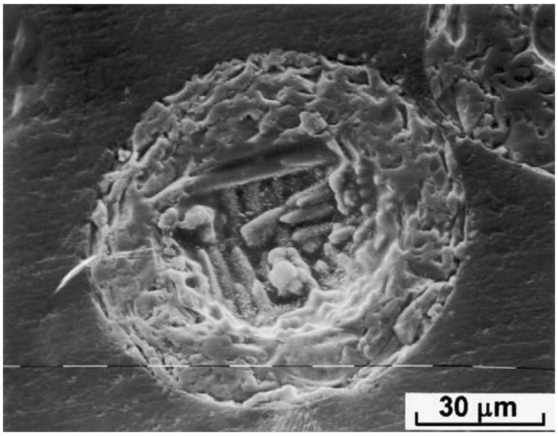

Fig. 8. Oxidized internal structure of a particle.

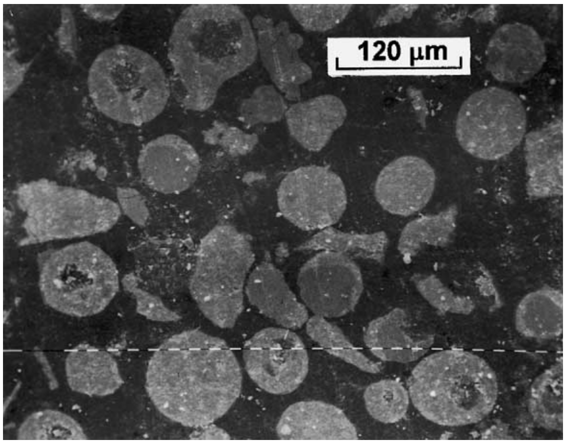

Fig. 9. Inclusions in the internal part of particles.

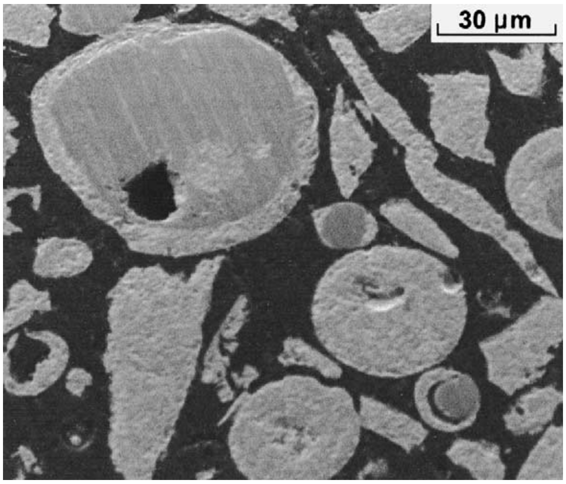

Fig. 10. Particle with nucleus of different composition than the external part, mostly Mo and U.

Table 2
EDS analysis of the dark zone in the center of the particles

Position	U (wt%)	Mo (wt%)
1	0	100
2	87.8	12.2
3	32.7	67.3
4	100	0
5	40.6	59.4
6	100	0
1	60.4	39.6
2	100	0
3	22.3	77.7
4	63.9	36.1
5	97.7	2.3

4. Discussion

The production of powders of different alloys can be obtained by different methods such as: the plasma rotating process, [11], and the gaseous vaporization already used to obtain U–Mo particles, [12]. These methods have in common with the electrodischarge the fusion of the material and the fast cooling rate of the liquid with the consequence of microsegregation problems. The first two conducted in an argon atmosphere, the electroerosion has the advantage that the particles can be mixed with components of the dielectric.

The electrosparked particles, according to the powder metallurgy shape characterization as depicted by ISO 3252, [13]. The particles are somewhat spheroidal with a smooth surface without satellites, some of them are nodular and others irregular. The spherical shape of the particles is explained taking into account the liquid state of the ejected material and its fast cooling during

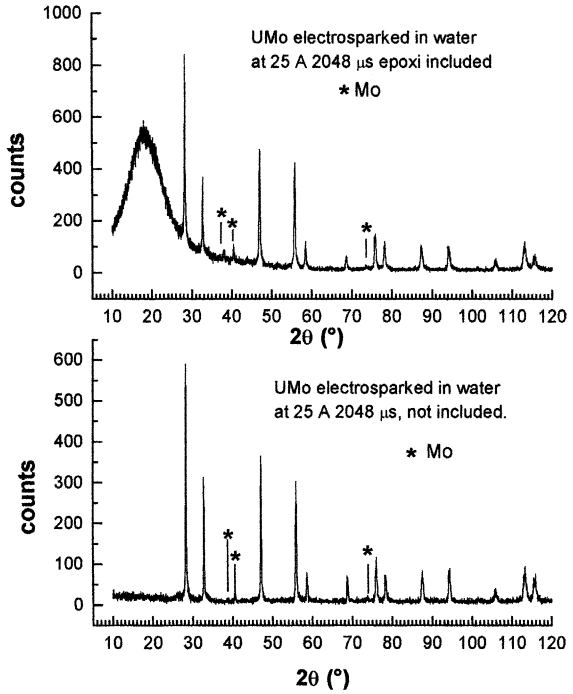

Fig. 11. Cu-Kα XRD of the particles, all lines correspond to UO_2 except the (*) marked.

its immersion into the dielectric, the surface tension and the energy minimum principle explain this specific geometry as occurs in many fields where drops are formed.

The nominal density of UO_2 is 10.91 $\mathrm{g\,cm^{-3}}$ so compared with the powder density measured of 9.86 ± 0.05 $\mathrm{g\,cm^{-3}}$ we can affirm that the mean value obtained corresponds approximately to UO_2 particles with holes and splits. The existence of metallic nucleus surrounded by a layer of oxides may be thought as if a chemical reaction of the alloy with the oxygen of the water were the fundamental process that took part during the liquid state of the material. The differences in composition in the nucleus are not related to the existence of different composition zones in the electrodes because dendrites did not appear in rods, so the different composition of the nucleus may be attributed to the difference in the melting point of the constituents of the ejected spheres.

In the case of using water as dielectric, for example separating the oxidized particles by densitometry, it would be useful to obtain UO_2 particles of different sizes.

Considering that the composition of the particles can be altered by the dielectric, or not if noble liquid gases are utilized, this process would be useful to obtain uranium carbides using kerosene instead of water as dielectric.

5. Conclusions

The method is interesting to nuclear applications to conform uranium powders composed with the elements of different dielectrics. EDM is a useful process to produce small somewhat spherical particles with sizes from few μm to near 100 μm.

Acknowledgements

E.D.C. is member of Consejo Nacional de Investigaciones Científicas y Técnicas of Argentina. L. Pérez for the density determinations, R. Fort and E. Bisi for collecting of particles.

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