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Effect of sodium injection rate in reduction process on characteristics of tantalum powders

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Abstract: The paper presents a research the effect of sodium injection rate in a melt containing potassium tantalum fluoride and a flux on morphology and characteristics of tantalum powders obtained by sodium reduction.

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1 Introduction

Solid electrolytic tantalum capacitors (SETC) are widely used in electronic technique. Their advantages over other capacitors are high specific charge per unit volume or mass, low leakage current and high reliability. Porous SETC anode sintered in vacuum from fine metal powder provides a high anode surface area. One of the widely used methods for capacitor power production is sodiumthermic reduction of tantalum in inert atmosphere from melts containing potassium tantalum fluoride and a flux. The powder surface value determines the specific charge while the metal's chemical purity determines the leakage current of capacitor. It is necessary to have pure reagents to obtain metals with low impurity contents but this, however, results in the formation of powders with insufficiently high specific surfaces. For increasing the specific surface of raw powders by change of particle growth conditions there is a usual practice to enter into the initial melt or into gas atmosphere reagent additives containing non-metallic impurities[1,2]. It is interesting search of others technological receptions to increase specific surface of raw powders in melts with minimum impurity level.

The objective of this work to study the effect of sodium injection rate upon melt surface during initial period of sodiumthermic reduction on characteristics of tantalum powders.

2 Experimental

Powders were produced by feeding liquid sodium onto the surface of a melt consisting of a tantalum salt and a diluent salt as a heat sink (liquid-phase reduction). As starting reagents K_2TaF_7 (every metal impurity content of less than 0.001 %(mass fraction)) and KCl (reagent grade) were used. The reduction was carried out in the temperature range of 700-800℃ at a unit whose installation diagram is presented in Fig. 1. Container 1 had the required quantity of sodium. The charge consisting of 2.0 kg K_2TaF_7 and 2.28 kg KCl (molar ratio 1:6) was loaded into a nickel vessel 8 and placed into a retort—reactor 10. After assembling the retort was placed into a furnace and heated in vacuum, gradually increasing the charge tempera-

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ture. When the temperature was 450℃ and pressure ~3 Pa, the retort was filled with argon, the salts were melted; then a stirrer 9 was lowered into the melt to mix it. Upon achieving the required temperature reactor was fed by liquid sodium along the pipeline 4 under a continuous stirring of the melt. After the reduction process was completed, the reactor was cooled to room temperature and metal powder was separated from salts by hydrometallurgical treatment described earlier[3]. The powder specific surface (S) was measured by the FlowSorb II 2300 installation. The bulk density (γ) of powders was determined by standard methods. For determining the electric characteristics of the anodes pellets from a powder without a binding agent with the diameter 2.95 mm and density 5.0g/cm^3 were compacted. The pellets were sintered in a vacuum resistance furnace at temperature 1400℃ and residual pressure not greater than 5×10^{-3} Pa. The exposure at a maximal temperature was 30 min. The anodes were oxidized at 80℃ in a 1% H_3PO_4 solution at a voltage of 70V.

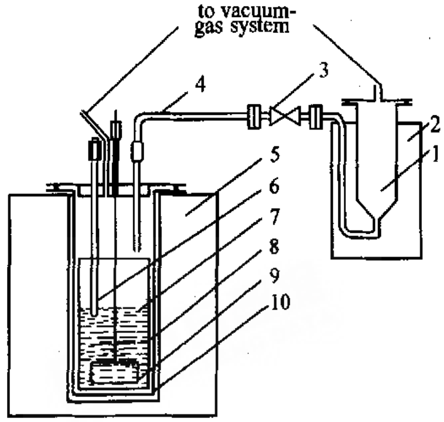

Fig. 1 Installation diagram of the experimental unit

1 — container with sodium; 2 — electric furnace of the sodium container; 3 — valve; 4 — pipeline; 5 — electric furnace of the reactor; 6 — thermocouple casing; 7 — melt; 8 — reaction vessel; 9 — stirrer; 10 — retort-reactor

3 Result and discussion

Before reduction process a molten bath of starting salts maintained at temperature above the liquidus temperature but significantly below the reduction temperature. Then melt is reacted with molten sodium until a required reduction temperature is reached. This period of the reaction during which the temperature of the molten bath increases from the initial temperature to the reduction one is called the "nucleation period"[4]. We carefully controlled the rate of sodium injection during this period. Some temperature—time diagrams in initial period of reduction at various sodium addition rates represented in Fig. 2.

Carried out experiments have shown that change of sodium injection rate and, accordingly, change of melt temperature represented in Fig. 2 noticeable influence on powder properties. This is reflected in fact that with other things being equals a change in granulometric composition and bulk density of produced powders takes place (Table 1).

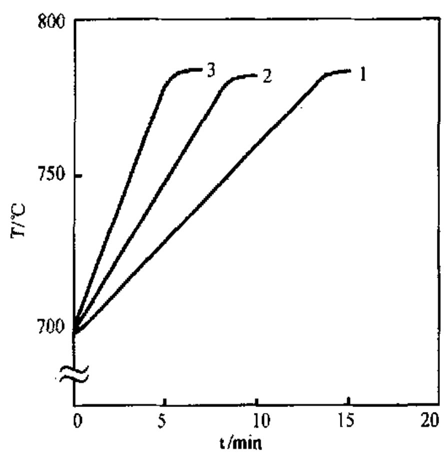

Fig. 2 Temperature — time diagrams in initial period of reduction at various sodium addition rates

1 — 10g/min; 2 — 15g/min; 3 — 20g/min

Table 1 Effect of sodium injection rate on granulometric composition and bulk density of tantalum powders

v /g·min^{-1}	The fraction content/%					γ/g·min^{-1}
	−50μm	50−71μm	71−100μm	100−200μm	200−400μm	
10	45.8	4.66	9.72	20.0	19.2	2.68
15	79.2	2.7	2.1	10.1	5.9	1.66
20	95.56	1.81	1.55	0.82	0.24	1.45

It is seen from Table that rate of sodium injection has an inverse effect on the particle size. As regards the powder morphology, it was established that the bulk of the powder mass, irrespectively of sodium injection rate, is represented by equal-axis nodular particles consisting of individual fragments. The fragments are connected by bridges. More rarely are particles shaped as regularly faceted crystals, which clearly exhibit growth stages. The nodular particles have the most developed surfaces. It is their contribution to the powder surface value that is the most appreciable. The growth of sodium injection rate during initial period of metal reduction enlarged the nodular particle fragments and, correspondingly, decreased their surface value. An increase in specific surface of powder has been stipulated by the rise in anode specific charge (Fig. 3)

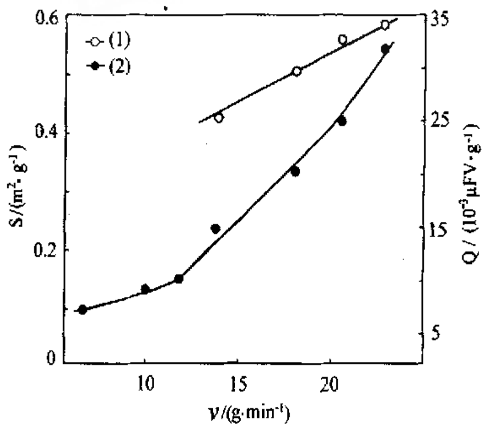

Fig. 3 Dependencies specific surface (1) of powders and specific charge of anodes (2) on rate of sodium injection

Thus, the research has shown that change of sodium injection rate renders essential influence to characteristics of tantalum powders.

4 Conclusion

The research has shown that temperature schedule during initial period of metal reduction seriously affects the process of powder particle crystallization and its grain sizes, respectively. The increase of sodium injection rate and, accordingly, the temperature change in initial period of reduction allows obtaining powders with more developed surface. The application of an optimal mode has allowed at use of the same reagents considerably to increase working characteristics of the raw powder.

References

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