



# Electroerosion dispersion, sorption and coagulation for complex water purification: Electroerosion waste recycling and manufacturing of metal, oxide and alloy nanopowders\*\*

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The application of EEDSC—electroerosion dispersion, sorption and coagulation (using *in situ* dispersion in plasma of aluminium or iron)—allowed efficient water purification from heavy metal ions and radioactive alkali ions (of Fe, Cr, Cu, Mo, Zn, Co, Ni, Cd, Mn, As, Sn, Pb, Al, Ba, Co, Cs and Sr) as well as from organic contamination. Intensive ozonization should be applied because of intensive foam formation as a result of complex electrohydrodynamic effects during removal of the “heavy” organic contaminants (such as from liquid waste from landfills) during EEDSC. The method of electroerosion dispersion is very effective for the production of nano and submicron powders (5 nm–3 µm) of metals, oxides, nitrides and carbides as well as for recycling of any conductive materials such as cemented carbides, alloys

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\*\* First presented at the Sustainable Industrial Processing Summit (SIPS)—Mamalis International Symposium, 4–7 November 2018 in Rio de Janeiro.

of heavy metals, metallic waste products etc. The iron oxide polyvalent magnetic nanoparticles produced by electroerosion dispersion attract considerable interest in many fields of research and application. They have high potential for applications in the field of biomedical sciences (diagnostics and therapy), ferrofluids, catalysis, coloured pigments, high-density magnetic recording, printer toners, Li-ion batteries, wastewater treatment and absorption of electromagnetic waves.

**Keywords:** aluminium oxides, electroerosion dispersion, iron oxides, nanopowders, sorption and coagulation, water purification

## 1. Introduction

Wastewater and drinking water treatment is a major problem. There are no universal methods of water treatment and each type of contaminated water needs a special approach. The known industrial manufacturing methods for nano and submicron spherical powders of refractory metals, hard and heavy alloys, metals having high plasticity, and chemically active, toxic and radioactive metals, are expensive, energy-consuming, environmentally hazardous and difficult to implement.

The processes of electroerosion dispersion (EED) and electroerosion dispersion, sorption and coagulation (EEDSC) using *in situ* dispersion in plasma of aluminium or iron granules or chips and equipment for their implementation have been patented by us in 2005–6 [1–4] and described [5–7]. The proposed EED method can be easily industrialized and gives the possibility of cheap and low power-consuming production of nano and submicron polydisperse powders of metals and their oxides, hydroxides, carbides and nitrides. They can be crystalline or amorphous, with particles of spherical or of arbitrary shape depending on the régime of EED. The aluminum and iron oxides and hydroxides can be used as sorbents and coagulants for waste and drinking water treatment—removal of ions of heavy, alkali, alkali earth and radioactive metals. EEDSC can yield inline production of iron or aluminum ions, hydroxides and oxides in wastewater and parallel destructive, deodorization and disinfection effects of plasma on contaminants in the treated wastewater.

Many researchers have studied the method of electrochemical coagulation for wastewater treatment [8–11]. But these methods are rather selective and do not have all the advantages of electroerosion dispersion, sorption and coagulation proposed by us [6]. Similar technologies were developed somewhat later [12–13]. Many different methods of water purification from radioactive and heavy metals and from organic contamination using membrane filters, sorbents and coagulants or ozonization (without the application of electric or electric discharge technologies) have been developed [14–43]. The proposed EEDSC method using *in situ* dispersion in plasma of aluminium or iron unites the different cleaning methods in a single process. The method is cheap, efficient, provides a high degree of purification and is ecologically friendly; the developed equipment for accomplishing EEDSC is very compact.

## 2. Experimental

The technological equipment for the energy-efficient production of nano and submicron powders (5 nm–3 µm) of metals, oxides, nitrides and carbides as well as for recycling of any conductive materials such as cemented carbides, alloys of heavy metals, any metallic waste

products etc. was developed, designed and produced by Monastyrrov (e.g., Figure 1). The units incorporate disintegration reactors and pulse generators, allowing varying discharge frequency (100–500 Hz) and voltage (100–500 V). The EED method is based on disintegration of metal granules or chips in liquids (water, alcohol, kerosene etc.) by metal evaporation in plasma generated by pulsed electric discharges between the metal granules or chips with subsequent condensation of vapour or microdroplets. Metal vapour and microdroplets almost immediately (in about  $10^{-6}$  s) solidify in the cold zone in the liquid. Despite the temperature in the places of plasma discharge being about 15 000 °C, the temperature of the walls of the reactor are near to room temperature. Metals may be oxidized by oxygen, which is produced in the plasma due to water decomposition, or may turn into carbides or nitrides if they are dispersed into carbon- or nitrogen-containing media. EED allows operation with temperature gradients up to  $\pm 10^6$  K/mm; as a result, micrograins form in distinctly nonequilibrium conditions. Such particle formation conditions result in many unique properties of the powder grains (including deformation of crystalline lattices and high specific surface of the grains).



Figure 1. EED dispersion installation: 1, generator; 2, reactor; 3, sedimentation tank; 4, pump for liquid.

EEDSC is in fact an EED method, but the dispersion of metal granules or chips is into contaminated water, which is pumped via the reactor, thus aluminum or iron is evaporated directly into the contaminated water under the influence of plasma, which appears between the granules or chips because of the generated electrical pulses.

The installation for intensive ozonization is shown in Figure 2a. The volume of the contaminated water tank is 2 m<sup>3</sup>. Ozone is generated *in situ* from the surrounding air by powerful pulses of current delivered via electrodes (Figure 2b).



Figure 2. Installation for the intensive ozonization of landfill water (a); and the process of ozone synthesis in a glow discharge (b).

### 3. Results and discussion

The processing line for EED dispersion with a capacity of 10 m<sup>3</sup>/hour can be located in an area of 36 m<sup>2</sup>, and with a capacity of 50 m<sup>3</sup>/hour for an area less than 80 m<sup>2</sup>. The compactness of the equipment is a big advantage for powder manufacturing in industrial volumes. The cost of electroerosion dispersion is about the same for all metals (10–12 USD per kg). Price varies depending on the preparation of granules, (some materials have to be chipped, others crushed). The technology allows recycling into powder of any conductive waste (metallic, hard alloys, heavy alloys, beer cans, chips, granules etc.). The specific energy intensity of the process is 1.5 to 3 kW for the production of 1 kg of powder. The process is cheap and ecologically friendly and there are no drains, no gas and no dust emissions. The EED method is most competitive in cases when high-quality powders are required (especially fine-grained, superclean, spherical or with a highly developed surface, etc.) and in the case of processing metals with extreme physical and chemical properties. The technology can be used for the industrial production of a wide range of high-tech competitive products, including nanostructured materials.

An X-ray study of dried EED powders showed that iron-based powder contains practically spherical particles of Fe<sub>3</sub>O<sub>4</sub> (mainly) and some FeO and Fe [6]. The aluminum-based powder after drying contained 92% of bayerite (Al(OH)<sub>3</sub>), 5.5 % corundum (α-Al<sub>2</sub>O<sub>3</sub>), less than 1% of gamma aluminium hydride (γ-AlH<sub>3</sub>), and about 2.4 % of aluminium (Al).

The EED-produced nanopowders (20–50 nm) of iron and aluminum oxides are effective sorbents and coagulants. Tables 1 and 2 present the results of cleaning water from heavy metal ions using dried EED aluminum- and iron-based oxides, which were mixed with contaminated water for 8–10 minutes. For optimal operation of the technology, the total concentration of heavy metal ions should not exceed 100 mg/L. If this limit is exceeded, cleaning does occur but the consumption of powder increases and the productivity of the equipment decreases; it is

better to first dilute the contaminated water to a concentration that will allow efficient sorption-coagulation using the proposed sorbents: wastewater treatment powder of iron oxides (consumption 6–7 grams of dry powder per gram of impurities). The quality of the treated wastewater corresponds to the requirements of the usual sanitary norms and state standards for wastewater treatment.

Table 1. Degrees of extraction of heavy metal ions from the galvanic drains of the radio engineering plant in Nikolaev (“Zarya”—“Mashproekt”), the Antonov aircraft plant, and the “Alkon-serviskomplekt” (acidic waste water) in Kiev, with EED-produced iron oxide powder.

Metal ion	Initial concentration /mg dm <sup>-3</sup>	Concentration after purification /mg dm <sup>-3</sup>
Al <sup>3+</sup>	16.2	<0.05
As <sup>3+</sup> and As <sup>5+</sup>	49.2	0.01
Cd <sup>2+</sup>	0.103	<0.001
Co <sup>2+</sup>	19.4	0.08
Cr <sup>6+</sup>	144.4	0.01
Cu <sup>2+</sup>	1.27	0.002
Fe <sup>2+</sup> and Fe <sup>3+</sup>	55.5	0.5
Mn <sup>2+</sup>	1.94	0.0006
Mo <sup>2+</sup>	8.71	<0.05
Ni <sup>2+</sup>	25.7	0.05
Pb <sup>2+</sup>	3.3	<0.1
Sn <sup>2+</sup>	6.9	0.1
Zn <sup>2+</sup>	100.2	0.36

Table 2. Degrees of extraction of heavy metal ions from the galvanic drains of the experimental plant of the Leningrad (St Petersburg) Radio Engineering Research Institute with EED-produced aluminium oxide powder.

Metal ion	Allowable concentration <sup>a</sup> /mg dm <sup>-3</sup>	1st test			2nd test		
		Initial concentration /mg dm <sup>-3</sup>	Concentration after purification /mg dm <sup>-3</sup>	% purification	Initial concentration /mg dm <sup>-3</sup>	Concentration after purification /mg dm <sup>-3</sup>	% purification
Al <sup>3+</sup>	0.02	16.2	< 0.05	99.7	13.0	<0.05	99.2
Cd <sup>2+</sup>	0.001	78	0.43	99.4	1.8	<0.01	99.4
Cr <sup>6+</sup>	0.03	234	0.07	100	180	<0.01	100
Cu <sup>2+</sup>	0.002	149.5	0.69	99.5	42.5	0.05	99.9
Fe <sup>2+</sup> and Fe <sup>3+</sup>	0.5	55.5	0.50	99.1	2.4	0.005	99.8
Ni <sup>2+</sup>	0.04	25.7	0.18	99.3	56	0.05	99.9
Pb <sup>2+</sup>	0.5	3.3	< 0.2	94.0	0.45	<0.03	93.3
Sn <sup>2+</sup>	—	6.9	0.09	98.7	4.4	<0.05	99.0
Zn <sup>2+</sup>	0.02	25.5	0.28	98.9	17.2	<0.005	100
Total	—	594.6	2.49	99.6	317.75	0.26	99.9

<sup>a</sup> According to sanitary norms and state standards for wastewater treatment.



Iron oxide-based sorbent can be successfully used for drinking water treatment (for removing of As, in particular). At the beginning, the drinking water should be passed through a self-cleaning filter (e.g., AMIAD) with mesh sizes of 10  $\mu\text{m}$  to remove sand. After As sedimentation by iron oxides the drinking water is ozonized in order to remove the dissolved Fe ions (which usually are present in the groundwater as well). In the final stage, the drinking water is passed through carbon filters.

The iron oxide polyvalent magnetic nanoparticles produced by electroerosion dispersion have considerable potential use in many fields of research and application. They can be used in the biomedical sciences (diagnostics and therapy), as ferrofluids, catalysts, coloured pigments, for high-density magnetic recording, printer toners, Li-ion batteries, wastewater treatment and absorption of electromagnetic waves [6].

Aluminum oxide slurry prepared by EED was delivered to the Chernobyl power plant (near Kiev, Ukraine) after storage for 60 hours and in the Chernobyl laboratory it was mixed with water contaminated by radioactive impurities. The results of the radioactive ion extraction can be seen in Table 3. Comparatively low coagulation and sedimentation of Cs ions can be explained by the decreasing activity of the aluminum-based oxide slurry.

Table 3. Results of a study in the Central Analytical Laboratory “Ecocenter” (Chernobyl, Ukraine) of radioactive ion extraction from solutions by mixing with EED-prepared aluminum oxide sludge (after more than 60 h since preparation).

Radionuclides	Radioactive solution №1				Radioactive solution №2				Radioactive solution №3			
	Initial activity /Bq dm <sup>-3</sup>	Final activity /Bq dm <sup>-3</sup>	$K_d^a$	BA% <sup>b</sup>	Initial activity /Bq dm <sup>-3</sup>	Final activity /Bq dm <sup>-3</sup>	$K_d^a$	BA% <sup>b</sup>	Initial activity /Bq dm <sup>-3</sup>	Final activity /Bq dm <sup>-3</sup>	$K_d^a$	BA% <sup>b</sup>
<sup>241</sup> Am	3.1	*		→ 100	31	*		→ 100	155	*		→ 100
<sup>60</sup> Co	1.08	*		100	10.8	*		→ 100	54	2.55±0.28	21	95
<sup>137</sup> Cs	1.62	1.2±0.48	1.4	26	16	10.1±0.51	1.6	37	80	48.2±1.3	1.7	40
<sup>90</sup> Sr	0.87	<MDA		→ 100	8.7	0.6±0.09	14.5	93	87	11.2±0.56	7.8	87

\* Was not detected.

<sup>a</sup> Decontamination factor  $K_d = A_i / A_f$ , where  $A_i$  and  $A_f$  are, respectively, initial and final activities.

<sup>b</sup> Percentage deactivation  $BA = [A_i - A_f / A_i] \times 100$ .

*In situ* experiments using model solutions (contaminated by nonradioactive isotopes of Cs and Sr)—prototypes of real (contaminated by the same elements, but their radioactive isotopes) of similar concentration in the EED reactor (or implementation of EEDSC technology) allowed us to essentially totally remove all contaminating ions. Dispersion in plasma-evaporated metal (Al) can pass through all valences before being finally oxidized and thus producing *in situ* sorbents-coagulants, which trap even ions having +1 (Cs) and +2 valences (Sr). As one can see from Table 3 even “old” EED-produced aluminium oxide-based sludge is able to effectively extract caesium-137 (<sup>137</sup>Cs), americium-241 (<sup>241</sup>Am), strontium-90 (<sup>90</sup>Sr), cobalt-60 (<sup>60</sup>Co), zinc (Zn) and lead (Pb) radioactive ions from polluted water.

Alongside the metallic ions in radioactive water, it is usually additionally polluted by organic contaminants. When a series of electropulse discharges passes through the water, powerful electrohydrodynamic shocks ensue with electromagnetic, ultrasonic and ultraviolet radiation pulses, and cavitation takes place, which effectively destroys all types of organic matter found in water in the form of oils, sodium ethylenediamine acetate etc. The complex effect on the water of such harsh physical agents engenders the destruction of the contaminating molecules, breaking chemical bonds. Chemical compounds that enter the zone of impact of the plasma discharge are reverted to the level of simple chemical elements with the subsequent reverse synthesis of compounds. Water is activated with the nucleation of  $\text{OH}^-$  radical ions, excited  $\text{H}_2\text{O}^-$ ,  $\text{H}_2$ ,  $\text{O}_2$ ,  $\text{H}_2\text{O}^+$ ,  $\text{H}^+$  ions and electrons  $\text{e}^-$ . The high local concentration of active particles determines their partial recombination and the synthesis of molecular products:  $\text{H}_2$ ,  $\text{H}_2\text{O}_2$ ,  $\text{H}_3\text{O}_2$ ,  $\text{H}_2\text{O}$ ,  $\text{O}_3$  etc. A distinctive feature of the process is that at the same time, particles with opposite properties are generated, providing oxidative ( $\text{OH}^-$ ,  $\text{H}_2\text{O}^-$ ,  $\text{H}_2\text{O}_2$ ,  $\text{O}_2$ ,  $\text{O}_3$ ) and reducing ( $\text{H}_2$ ,  $\text{H}_2\text{O}^+$ ,  $\text{H}^+$ ) reactions.

Unfortunately, the EEDSC method is inapplicable to landfill water contaminatet with “heavy” organic matter. The destruction of organic contaminants occurs so efficiently that large amounts of foam are formed (Figure 3), preventing effective cleaning, while the intensive ozonation is of great help using the installation shown in Figure 2. Using this installation,  $1 \text{ m}^3$  of heavy organic-polluted water with a chemical oxygen demand (COD) of  $9000 \text{ mg O}_2/\text{dm}^3$  can be cleaned in 24 h (using 79.5 kWh of electricity) resulting in  $16 \text{ mg O}_2/\text{dm}^3$ , which is 10 times lower than the safe limit.



Figure 3. The foam formed as a result of destruction of heavy organic contaminants.

#### 4. Conclusions

The presented technologies of electroerosion dispersion (EED) of powder production and electroerosion dispersion, sorption and coagulation (EEDSC) for water purification have a wide spectrum of applications, are very efficient, cheap and ecologically friendly. The technology of intensive ionization mimics natural processes of water cleaning, is very efficient for drinking water treatment and helps to solve the problems with sewage water. The level of technological development allows us to design and manufacture industrial equipment for particular needs.

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