

PREPARATION OF RADIOACTIVELY LABELLED CONDENSATION AEROSOLS. IV.*

AEROSOLS OF GOLD, RHENIUM OXIDE, SILVER, TELLURIUM AND VANADIUM OXIDE

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Received April 1st, 1970

The reasons for preparation of radioactive labelled aerosols and the advantages found in their application have been thoroughly described in the preceding communications^{1,2}. The spontaneous condensation of saturated and supersaturated vapours of metals and of some inorganic compounds proved to be a good method for preparation of model aerosols³⁻⁶. The formation of a condensation aerosol cannot be described and predicted perfectly by using a theoretical treatment, only. The applicability of the classical or even the statistical theory of vapour condensation makes it not possible till now^{5,6}. Of course some of qualitative theoretical conclusions are useful for experimental purposes. The whole formation process of condensation aerosols is often given by a superposition of three different processes, *i.e.* nucleation, nuclei growth by condensation and particle coagulation. There are also practical difficulties in the use of the classical theory, which are due to the insufficiency or absence of data of the vapour pressure of many substances and of their surface tension data.

The methods described in this paper offer approximately monodisperse aerosols prepared by a spontaneous vapour condensation in the absence of condensation nuclei. The preparation

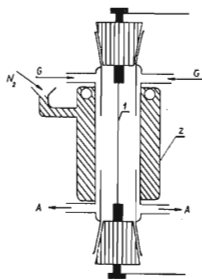


FIG. 1

Apparatus for Preparation of Condensation Aerosols by Heating of Metal Wires

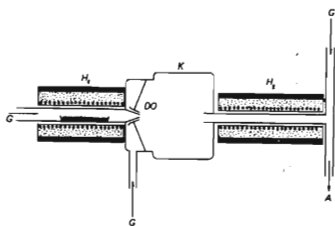


FIG. 2

Apparatus for Preparation of Concentration Aerosols by Means of Double Furnaces

* Part III: This Journal 36, 2368 (1971).

takes part either in furnace generator or in a heated wire generator. The radioactive labelling of aerosols is done by means of radioactive labelled elements and compounds.

EXPERIMENTAL

Apparatus

For practical purposes it seems to be necessary to find — using those qualitative conclusions of the theory — some optimum reproducible conditions for vaporizing of metals and inorganic compounds as well as for the condensation of these vapours. Two methods proved to be most suitable for these purposes: spontaneous volume condensation of metal and metal oxide vapours (heated wires) or spontaneous condensation of vapours in a free turbulent jet⁶.

Both methods are being used for preparation of radioactive labelled inorganic aerosols (Fig. 1 and 2).

In the first case a wire is heated electrically and evaporized. Its vapours are due to form nuclei and then aerosol particles near the cold walls of the glas tube. In our investigations this generator (Fig. 1) was used for preparation of Re_2O_7 aerosols by heating a rhenium wire (1). The walls 2 of the glas generator were cooled by means of water or by means of liquid nitrogen. The wires of gold and silver were not suitable for this generator, they melted before their vaporization. Clean dried air G was flowing laminarly along the walls. The condensation aerosol A was led for sampling and radiochemical analysis.

The furnace generator (Fig. 2) is more suitable for vaporizing of metals and some inorganic compounds of lower m.p. and for following condensation of their vapours. The substance is

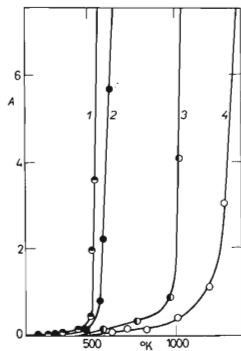


FIG. 3

Aerosols Concentration (radioactivity A) as a Function of Vaporizing Temperature
1 Te, 2 Re_2O_7 , 3 Ag and Au, 4 V_2O_5 .

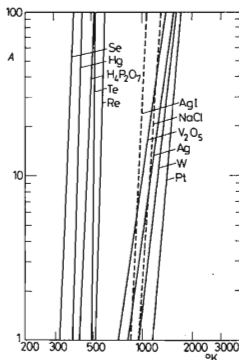
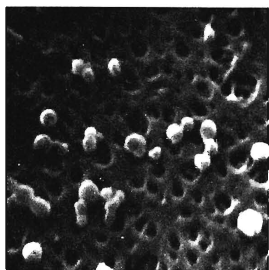


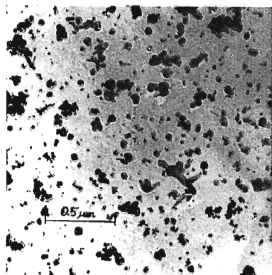
FIG. 5

Aerosol Concentration (radioactivity A) as a Function of Vaporizing Temperature

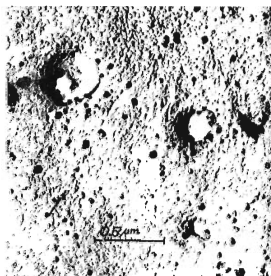
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a



b



c

FIG. 4

Micrographs of Aerosol Particles (Replica — technique, the surface of a membrane filter)

a Re_2O_7 , particle size $2\bar{r} \approx 0.05 \mu\text{m}$, vaporizing temperature 330°C ; *b* particle size $2\bar{r} \approx 0.041 \mu\text{m}$, vaporizing temperature 270°C ; *c* Ag, particle size $2\bar{r} \approx 0.038 \mu\text{m}$, vaporizing temperature 650°C .

TABLE I
Radioactively Labeled Inorganic Condensation Aerosols

Compound or element ^a	Temperature range, °C	Range of particle radii, μm	Radioactive isotopes
Pt-oxides, mo, s	600—1 300	$5 \cdot 10^{-3}$ — $3 \cdot 10^{-2}$	¹⁹⁷ Pt (18 h) ¹⁹⁹ Au (3 d)
Ag, s	600—1 100	$2 \cdot 10^{-2}$ — $2 \cdot 10^{-1}$	¹¹⁰ Ag (249 d)
Au, s	700—1 200	10^{-2} — 10^{-1}	¹⁹⁸ Au (2.7d)
WO ₃ , N ₂ , mo, s	900—1 200	$2 \cdot 10^{-2}$ — $8 \cdot 10^{-2}$	¹⁸⁵ W (73d)
NaCl, mo, s	400—1 100	$3 \cdot 10^{-3}$ — 10^{-1}	²⁴ Na (15 h) ²² Na (2.6 years)
V ₂ O ₅ , s	400— 950	$5 \cdot 10^{-2}$ — $1.5 \cdot 10^{-1}$	⁵⁰ V (6.10 ¹⁴ years)
Se, N ₂ , mo, s	150— 300	$3 \cdot 10^{-2}$ — $3 \cdot 10^{-1}$	⁷⁵ Se (27 d)
Te, N ₂ , mo, s	200— 300	10^{-2} — $5 \cdot 10^{-2}$	¹²⁷ Te (105 d)
Re(Re ₂ O ₇), s	100— 350	$4 \cdot 10^{-3}$ — $3 \cdot 10^{-2}$	high spec. act. ¹⁸⁶ Re (90 h) ¹⁸⁸ Re (17 h)
AgI, N ₂ , mo, s	200— 600	$5 \cdot 10^{-2}$ — $3 \cdot 10^{-1}$	¹³¹ I (8 d) ¹¹⁰ Ag (249 d)
H ₄ P ₂ O ₇ , s	150— 300	10^{-2} — 10^{-1}	³² P (14 d)
H ₂ SO ₄ , l	50— 200	10^{-1} — 10^0	³⁵ S (87 d)
Hg, N ₂ , l	50— 200	$5 \cdot 10^{-1}$ — 10^1	²⁰³ Hg (48 d)

^a mo means monodisperse aerosol, s solid, l liquid.

heated in a furnace H_1 and its vapours form aerosol in a double jet DO by mixing of vapours and cold gas (chamber K). A second furnace (reheater H_2) can be used for a resublimation of primarily formed aerosols. The second condensation is carried out by mixing of clean gas G with a vapour stream. This "double" condensation method have made the width of the particle size distribution curves narrower. In this generator the aerosols of gold, silver, tellurium and vanadium oxide (V_2O_5) were prepared.

Materials

Radioactive labelled gold p.a. purity showing a specific activity of about 6 Ci/g was used. It was the isotope ¹⁹⁸Au (2.7 d). For the aerosol preparation the quantity of 25 mCi was used, which was added into 0.5 g of non-activated gold. The preparation of aerosol was carried out in the apparatus according to Fig. 2.

Radioactive labelled rhenium. A rhenium wire of a high purity grade was applied, having the diameter of 0.2 mm and the length of 60 mm. The wire was radioactively labelled with the isotopes ¹⁸⁶Re (90 h) and ¹⁸⁸Re (17 h). The total activity of the wire was about 50 mCi and the specific activity about 5 Ci/g. For the aerosol preparation the apparatus according to Fig. 1 was used.

Radioactive labelled silver. About 1 mCi of radioactively labelled metallic silver of p.a. grade was inserted into the apparatus according to Fig. 2. Then it was added about 0.5 g of non-activated silver. Radioactive labelling was carried out with the isotope ^{110}Ag (249 d). The specific activity was about 100 mCi/g.

Radioactive labelled tellurium. About 10 mCi of radioactively labelled metallic tellurium (~ 10 mg) of p.a. grade was inserted into the apparatus according to the Fig. 2. Radioactive labelling was carried out with the isotope $^{127\text{m}}\text{Te}$ (150 d). The specific activity was about 1 Ci/g.

*Radioactive labelled vanadium.** About 0.02 g of V_2O_5 with a stable isotope ^{50}V was added into 0.5 g of natural V_2O_5 (p.a.). The concentration of ^{50}V ($6 \cdot 10^{14}$ years) was about 23%. For aerosol preparation the apparatus according to Fig. 2 again used.

Sampling and Measuring

The sampling of generated aerosols for measurement of radioactivity and particle size was made by means of Nuclepore and membrane filter technique⁷. After the measuring of radioactivity the same filter had been used for investigation of particles by means of electron microscopy. About 500 particles were measured in each sample. Then the distribution curve of particle radii was plotted and the mean arithmetic particle radius (r) and the Fuchs' factor α ($\alpha = \sigma/\bar{r}$; σ is the mean quadratic deviation of the radii) were calculated. According to Fuchs⁸ such an aerosol may be considered monodisperse, in which $\alpha \leq 0.2$.

RESULTS AND DISCUSSION

We have not been able yet to measure the partial pressure of these inorganic vapours and we could not know the supersaturation degree. Therefore we plotted the weight concentration (radioactivity) of generated aerosols as a function of vaporizing temperature, only (Fig. 3). Similarly it was possible to calibrate the mean particle size as a function of vaporizing temperature and gas flow velocity (through the equipment)^{1,2}. The calibration, particle size as a function of vaporizing temperature at a constant gas flow was done by means of electron microscopy method (Fig. 4a,b,c**).

The gold and silver aerosols could be prepared by vaporizing of these metals at temperatures 900–1300°K (m.p. of Ag is 960.8°C and m.p. of Au is 1063°K) with particle sizes about 0.02 to 0.2 μm diameter. Rhenium metal heated to 370–600°K (m.p. 3180°C) by air flow of 0.5 l/min gave a Re_2O_7 -aerosol with particle sizes of 80–600 Å diameter. Rhenium can be obtained (like Au) in high specific radioactivities — about 5 Ci/g — and therefore aerosol concentration of about $10^{-4} \mu\text{g}/\text{m}^3$ is measurable. The tellurium aerosol could be prepared by vaporizing of metallic Te at temperatures 480–560°K (m.p. 449.5°C) with particle sizes of about 0.1–0.3 μm diameter.

As far as the degree of monodispersity α concerned, in most cases it was found that $\alpha > 0.2$, except at the aerosol of tellurium ($\alpha \sim 0.18$). The electrostatic particle charge of these aerosols was not measured.

From the results obtained up to the present it follows that the method of controlled vapour condensation is suitable for the preparation of radioactive labelled aerosols. It is possible to choose elements and inorganic compounds with suitable properties for the preparation of good model

* Radioisotopes were imported from the USSR (Re and V) and from German Democratic Republic (Ag, Au and Te).

** See insert on p. 3362.

aerosols. Till now 13 different inorganic aerosols have been already described^{1,2} (see Table I and Fig. 5). The most suitable for laboratory investigation in aerosol physics and chemistry are aerosols of Pt-oxides, Ag, Au, WO₃, NaCl, Se, Te, Re₂O₇ and AgI.

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REACTION OF POTASSIUM CARBONATO-BIS(GLYCINATO) COBALTATE III WITH (S)-(+)-GLUTAMIC ACID; ISOLATION OF WATER SOLUBLE PRODUCTS

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Received September 29th, 1970

The expulsion of the carbonate group from the racemic [Co(en)₂CO₃] (ClO₄) (where en is ethylenediamine) by (S)-(+)-glutamic acid has the stereospecific formation of Λ-[Co(en)₂(S-(+)-Glu)] · (ClO₄)¹ as a consequence. The stereospecificity, consisting in the interaction of the γ-carboxyl with the NH₂ group of ethylenediamine, is of a kinetic character¹. Similarly, the CO₃²⁻ group in K[Co(AB)CO₃] (where AB is glycine or (S)-valine) also can be expelled. This reaction with (S)-alanine, (S)-valine, and (R)-valine was employed by Shibata and coworkers² for the preparation of mixed chelates of amino acids of the [Co(AB)(A'B')₂] or [Co(AB)₂(A'B')] type. The reaction gives both *cis* and *trans* isomers. In the case of (S)-leucine we have shown³ that the reaction leads to a mixture of all mixed forms, of both *cis* and *trans* isomers. A special position of glutamic acid with respect to the γ-carboxyl led us to an investigation of the reaction products of racemic K[Co(Gly)₂CO₃] with the mentioned acid.

The reaction with (S)-(+)-glutamic acid ([α]_D +30° in conc. hydrochloric acid) was studied under the conditions used by Shibata and coworkers². The course of the reaction was followed by paper electrophoresis. After filtering off the insoluble products from the reaction mixture (probably *cis* isomers) we proved the presence of four substances with different mobility, but all four moved to the anode (Table I). Therefore we separated the reaction mixture further on a pre-