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BAŞBAKANLIK
ATOM ENERJİSİ KOMİSYONU**

ÇEKMECE NÜKLEER ARAŞTIRMA VE EĞİTİM MERKEZİ

RAPOR No: 101

**CHEMICAL DOSIMETERS IN AQUEOUS
SOLUTIONS**

A. TUĞSAVUL and E. ÜNSEREN

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INTRODUCTION

The aim of this work was to describe the development of simple chemical dosimeters in aqueous solutions which would not require complicated equipment and rare, expensive initial materials.

The selection of a radiation sensitive system for dosimetric purposes involves a number of problems. In order for a system to be used as a dosimeter in a specific range, it should be sensitive to the reaction due to irradiation. System should have energy and dose rate independence, stability toward light and heat. The amount of chemical change should be directly proportional with dose. Besides of these the analytical method used to measure the dose from radiation absorbed system must be simple and convenient. Small amounts of impurities cause large deviations in the proportionality between chemical change and dosage especially in aqueous systems and some stabilizers must be added. In some cases sensitizers can increase the reproducibility of the reactions.

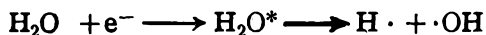
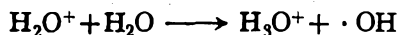
This review covers on dose determinations for electrons, γ - and X-rays and also pile radiations. It was divided into two parts as inorganic and organic chemical dosimeter systems.

RADIOLYSIS OF WATER

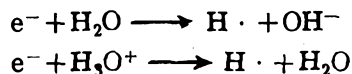
The radiolysis of water is complicated and over-all radiation process can be separated into several steps. The primary reaction under the ionizing radiation is assumed to be:



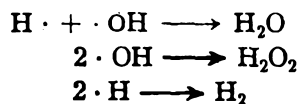
The product H_2O^+ either reacts with water or recaptures electron and dissociates to H and OH radicals,



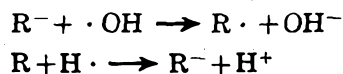
also the exited electron reacts with water and H_3O^+



In case of irradiation with heavy particals the radicals have sufficient density to combine locally to form H_2 and H_2O_2 but for electrons and γ -radiations main effect is the radical attack.



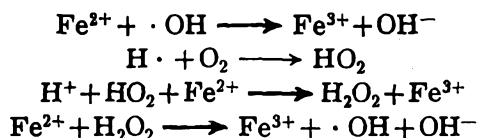
Small amounts of impurity present in water causes decomposition and radiation products increase in the solution.



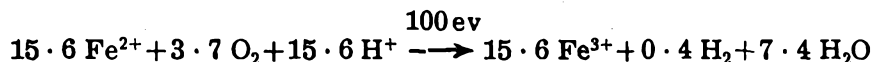
INORGANIC CHEMICAL DOSIMETERS

FeSO_4 (Fricke) Dosimeter

FeSO_4 in diluted sulfuric acid is the most widely used aqueous dosimeter and has been studied thoroughly. It comes very close to satisfy all the requirements. The reaction mechanism can be explained by the formation of H and OH radicals.



The over-all process can be explained in one equation:



The yield of ferric ions, number of molecules reacting per 100 ev, has been found by Hardwick (1) as 20.8 ± 0.3 for Co-60 γ -rays. C. J. Hochanadel and J. A. Ghormly (2) have determined the yield of FeSO_4 for γ -rays in air saturated 0.4 M sulfuric acid solutions and found

15.6 ± 0.3 . This result agrees with the value obtained by R. M. Lazo et al. (3) but it is 30 % lower than the result given by S. Davison et al. (4). Air saturated FeSO_4 solutions were irradiated by electrons and the yield has been found as 15.6 ± 0.5 in the 1-2 MeV energy range (5). Different G values have been determined by fission fragments for the oxidation of air equilibrated FeSO_4 solutions. It is concluded that the G value is a function only of the linear energy transfer (6) and is independent of the radiation intensity (7).

N. Miller (8) has showed that the oxidation rate was independent of the ferrous ion concentration from 10^{-4} M to 10^{-3} M and also independent of the sulfuric acid concentration above 0.1 N (9). J. Weiss (10) indicated that the oxidation of FeSO_4 is dependent to the oxygen concentration dissolved in solution. The addition of NaCl to the Fricke dosimeter has been recommended by J. Weiss et al. (11). Chloride ions protect the oxidation of Fe^{2+} by organic impurities and eliminate the requirement of very pure chemical reagents and water. W. G. Rothschild and A. O. Allen (12) have found that Fe^{3+} concentration can be increased by lowering pH and by adding NaCl.

The sensitivity of the Fricke dosimeter has been tried to improve by the application of more suitable chemical measuring methods. Addition of butyl alcohol increases the rate of formation of Fe^{3+} and this can be determined spectrophotometrically with CNS^- at 480 nm (13). In this case the lower limit of the Fricke dosimeter can decrease from 4.10^3 to 100 rads. The oxidation rate of Fe^{2+} ions increases in the alcoholic medium and the presence of NaCl in sufficient concentration suppresses the effect of the alcohol. The lower limit of measurability can decrease to a few hundred rads in Fricke dosimeter if ferric ion concentration is determined by KCNS (15) colorimetrically.

G. Rudstam and T. Svedberg (16) have used Fe-59 as a tracer for the determination of Fe^{3+} ions in Fricke dosimeter, between 0-100 rads dose range within two percent deviation. After irradiation, ferric ions were extracted from ferrous ions by isoamyl alcohol as ferric thiocyanate. Determination of doses in the range of 40-1000 rads with the Fricke dosimeter using radioactive iron has been improved by O. S. Gal (17).

S. S. Kuznetsova and I. K. Sokolova (18) investigated the effect of benzene as a sensitizer on the Fricke dosimeter. The magnitude of $G(\text{Fe}^{3+})$ increased from 15.68 to 53 ions per 100 ev in the presence of benzene in the system.

Addition of aromatic acids especially benzoic acid to the FeSO_4 dosimeter has an effect on the G value and doses of 3000 rads can be measured with sufficient accuracy (19).

FeSO_4 dosimeter has been modified to extend the upper usable limit of the dose by the addition of CuSO_4 . This modification has been investigated by E. J. Hart and P. D. Walsh (20). H. Hotta et al. (21) have also improved this CuSO_4 addition. Cu^{2+} ions cancel the effect of oxygen on the FeSO_4 dosimeter and the system serves as an accurate chemical dosimeter for high dose range of γ -rays.

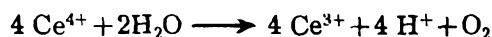
After irradiation of FeSO_4 solutions the quantity of unoxidized Fe^{2+} has been determined spectrophotometrically, based on the red color of the complex formed with orthophenanthroline (22). H. Bouziques et al. (23) have proposed this ferrous orthophenanthroline method for the determination of medium doses.

Fricke dosimeter containing Li_2SO_4 in various concentrations has been suggested by I. Draganic and J. Sutton (24) as a specific dosimeter for thermal neutrons in the presence of other ionizing radiations. J. Wright (25) has concluded that among aqueous solutions of FeSO_4 , benzene, sucrose and mandelic acid the Fricke dosimeter provides more suitable monitor for the pile radiations.

The sensitivity of the FeSO_4 dosimeter can be increased by the determination of Fe^{3+} concentration spectrophotometrically at a wavelength of 224 nm instead of at 304 nm (26). The molar absorptivity (ϵ) of Fe^{3+} ions in 0.8 N sulfuric acid measured in the 305 nm region differs by as much as 6 %. The most probable value of (ϵ) at 305 nm is assumed to be $2127 \text{ mol}^{-1}\text{cm}^{-1}$ using the same set of solutions and different spectrophotometers by T. H. E. Bryant and T. P. Ridler (27). Statistical analysis showed that errors in Fricke dosimeter arise from both the preparation of the reference solutions and the spectrophotometer used. It is concluded by R. Broskiewicz and Z. Bulhak (28) that iron dissolved in sulfuric acid gives more reliable results than iron salts. Intercomparisons have been carried out between five laboratories. The differences in between Fe^{3+} yields do not exceed 0.5 %. Fe^{3+} molar absorptivity (ϵ) was also intercompared in these five laboratories, and significant differences were found between independent determinations and concluded that the measurement of (ϵ) is the main source of the over-all uncertainty in the FeSO_4 dosimeter (29).

Ce(SO₄)₂ Dosimeter

The most important dosimeter other than Fricke is the ceric dosimeter which is based on the reduction of ceric ions to cerous by the decomposition of water under the irradiation. The over-all process is:



Ceric dosimeter can be used to measure doses up to 10⁸ rads, which in this dose range Fricke dosimeter is not applicable.

S. I. Taimuty et al (30) have prepared the ceric sulfate solution in the 0.001-0.05 M concentration range. Dilution has been carried out after irradiation, by 0.8 N sulfuric acid and the optical density was measured at 320 nm. J. T. Harlan and E. J. Hart (31) concluded that concentrated Ce(SO₄)₂ solutions are suitable for the measurement of the doses up to 10⁸ rads with a precision of $\pm 2\%$. The system shows energy independence in 10⁵–10⁷ rads dose range (32).

The yield of cerous ions reduced per 100 ev was found 2.52 ± 0.05 by C. J. Hochanadel and J. A. Ghormly (2). T. J. Hardwick (33) has found that 3.30 ± 0.03 ceric ions reduced per 100 ev for X- and γ -radiations. The yield is independent of ceric and cerous ion concentration in a specific range. The lower limit of this range is 2.10^{-6} M. When Ti₂SO₄ was added (34) to the ceric sulfate dosimeter the yield was increased from 2.39 to 7.92.

The yield was decreased from 2.79 to 2.54 by the addition of 0.001 M CuSO₄ to the ceric dosimeter but the presence of copper ions eliminate the requirement of the triple distilled water (35).

Ceric sulfate in borate solution (36) is a suitable system for the determination of neutron dose, γ -dose and heavily ionizing radiation in nuclear reactors. J. Preve and G. de Gaudemaris (37) have concluded that ceric and cyclohexane were the most reliable and reproducible dosimeter systems for the measurement of accelerated neutron radiations.

Ceric system is extremely sensitive to the impurities and light. The effect of light on ceric solutions was investigated by S. W. Nicksic and J. R. Wright (38) for the different concentrations and they found that sensitivity increases in diluted solutions. H. Hotta and K. Shimada (39) have observed slight temperature dependence of ceric sulfate system between 20°-90° C range.

K₂Cr₂O₇ Dosimeter

Potassium bichromate has been investigated as a high level γ -dosimeter system. Bichromate dosimeter is usable for absorbed doses between 10^5 and $3 \cdot 10^6$ rads range and permits the determination of γ -fluxes up to $5 \cdot 10^7$ rads/hour. 0.01 N solution of potassium bichromate in 0.8 N sulfuric acid has been irradiated (40) under the ionizing radiation. Color change has been observed because of the reduction of Cr^{6+} to Cr^{3+} and the optical density was measured at 350 nm. T. Asai and S. Ikegucki (41) have obtained reliable results in the dose range of 10^4 to 10^7 rads with a yield of 0.446 ± 0.023 . The reaction is proportional to the radiation dose but independent of dose rate. G. C. de Baere et al. (42) showed that although the yield is independent of the dose rate, it is influenced by the H^+ concentration. The reduction process is considerably effected by the pH of the solution (43). The oxidation and reduction processes reach in equilibrium above pH 4 and the concentration of Cr^{6+} remains unchanged under the action of radiation.

OTHER INORGANIC AQUEOUS DOSIMETER SYSTEMS

The aqueous solution of ferrine (ferric - 1, 10 - phenanthroline) is reduced to ferroin under the radiation and this process has been employed for the γ -ray dosimeter of the pile radiations. The yield equals to 12.5 for γ -rays (44). Yamaguchi et al. (45) have used aqueous solution of Mohr salt as a γ -rays dosimeter, measuring the amount of precipitation of Fe_3O_4 produced under the radiation. The volume of precipitation is a function of radiation dose.

Bariumplatinocyanide solutions were irradiated with γ -rays and the optical densities of the irradiated solutions were measured at the maximum absorption at 216 and 255 nm. The decrease of absorption is proportional with the dose of radiation until a total dose of 10^6 rads is reached (46).

Oxidation of stannous chloride in aqueous hydrochloric acid solution has been proposed as chemical dosimeter (47). T. Shiokawa and K. Hasegawa (48) have suggested titanium sulfate solution for the γ -rays dosimeter. The color produced after irradiation of acidic Ti-sulfate solutions, and the absorbance of the irradiated solutions at 410 nm, is proportional to the dose in the 10^5 - 10^8 rads range. 0.002 M Ti-sulfate in 0.8 N sulfuric acid has been used and a yield equals to 1.03 at 20°C has been found. The yield can increase to 4.19 by the addition of benzene to the solution.

The yield of bromate and bromide were measured in γ -irradiated alkaline hypobromide solutions. $G(\text{BrO}^-)$ was independent of temperature, dose rate and base strength between the range 0.01-0.1 M NaOH. It was concluded that the alkaline hypobromide system is suitable for chemical dosimetry (49).

Alkali nitrate solutions were irradiated with X-rays and it was found that nitrite produced from alkali nitrate solutions is proportional to the radiation absorbed. $\text{Ba}(\text{NO}_3)_2$ and BaCl_2 additions increase the production of nitrite (50). Decompositions of aqueous solutions of KNO_3 and NaNO_3 were investigated as a chemical dosimeter by G. Orban et al., (51). The nitrite ions produced by the irradiation were determined measuring the absorbance at 520 nm with addition of Griess-Ilosvay reagent to the irradiated solutions. It was concluded that KNO_3 solution in 2.5 M concentration was suitable as dosimeter in the 80 KeV-1.3 MeV energy range.

Reduction of chloroauric acid solution has been utilized by H. Fujita et al. (52). The yield of reduction of Au(III) has been determined by conductometry and found a value of 2.13. The system is not dependent on oxygen concentration, dose rate and temperature.

Ammonium molybdate dosimeter (53) can be used in the range of 10^4 - 10^7 rads for Co-60 sources. The reduction of $(\text{NH}_4)_2\text{MoO}_4$ is proportional with dose and can be measured either by titration with a standardized KMnO_4 solution or spectrophotometrically at 960 nm.

ORGANIC CHEMICAL DOSIMETERS

$\text{H}_2\text{C}_2\text{O}_4$ Dosimeter

It has been observed in the irradiated $\text{H}_2\text{C}_2\text{O}_4$ solutions that there is a definite relation between the amount of oxalic acid decomposed and the total dose absorbed (54). I. Draganic (55) has suggested oxalic acid system for dosimetry purposes between 1.6 and 160 megarads range. The yield for decomposition of $\text{H}_2\text{C}_2\text{O}_4$ is independent of temperature, radiation intensity, concentration of the solution and it is constant up to about 30 % decomposition. An oxalic acid chemical dosimeter has been used by A. L. Glass (56) to determine high level γ -radiation doses in the range of $1.6 \cdot 10^6$ - $6 \cdot 10^7$ rads. The oxalic acid system is not made radioactive by neutrons, making it suitable for in pile radiation measurements (57). The system is neither sensitive to impurities nor photosensitive.

$\text{H}_2\text{C}_2\text{O}_4 \cdot 2\text{H}_2\text{O}$, $\text{H}_2\text{C}_2\text{O}_4$ and other oxalate complexes (58) were irradiated with Co-60 γ -rays and evolved gases were analysed by mass spectrometry. The quantities of decomposed complexes were determined by spectrophotometry. G. Z. Gochaliev and Ts I. Zalkind (59) have investigated hydrogen formation from the oxalic acid solution in 0.8 N sulfuric acid and found that the rate of hydrogen formation is a linear function of the γ -radiation dose rate. Hydrogen concentration is maximum in between $3 \cdot 10^{-3}$ and $2 \cdot 10^{-2}$ N oxalic acid solutions. K. Sehested et al. (60) have determined the absorbed dose in oxalic acid solutions calorimetrically. The remaining $\text{H}_2\text{C}_2\text{O}_4$ has been measured by volumetric titration with KMnO_4 within an error of approximately 5 %. I. Draganic (61) have suggested NaOH titration to determine oxalic acid concentration after removal of CO_2 from the irradiated solution which would minimize the deviation due to chemical yield variation. When an adequate initial concentration is applied and the irradiation field is reasonable homogeneous the reproducibility of the oxalic acid dosimeter is almost perfect.

CHLORAL HYDRATE DOSIMETER

A chloral hydrate agar gel system was described by H. L. Andrews et al. (62) for use as a chemical dosimeter. HCl, formed from the chloral hydrate, has been determined either spectrophotometrically when an indicator such as methyl-red was added or by measuring the pH of the solution. Amount of acid formed from the molar aqueous chloral hydrate solution varied linearly with dose at a given dose rate (63). The change of conductivity (64) of the irradiated chloral hydrate solutions has been measured and found to be linear with dose and independent of dose rate. K. Wolschendorf (65) has also investigated the electrical conductivity changes of aqueous solutions of chloral hydrate. It was concluded that the system was operative at doses as high as 30.000 rads/min. The yield of HCl is independent of dose rate but depends on the initial concentration of chloral hydrate (66). R. F. Platford and J. W. T. Spinks (67) have observed a large dose rate dependent G value which is equal to 965 for the beta rays. R. M. Dziak (68) has showed that the yield is dependent of dose rate below 50 rads/min. for the γ -rays, thermal and fast neutrons. The temperature dependence has been investigated by several authors. Addition of 0.025-0.1 % resorcinol (68) eliminates the variations of the response with the temperature. Aqueous chloral hydrate with a pH indicator

dosimeter system has been suggested by P. M. Quiriconi (69) to measure the doses in a range between 100 to 500 rads. This system has limitation below 50 rads. It is concluded that chloral hydrate solution can be useful for low dose chemical dosimetry. The system is sensitive toward sunlight and the solution must be freshly prepared before irradiation.

HALOGENATED HYDROCARBON DOSIMETERS

A suitable dosimeter system has been suggested by J. W. Kanwisher (70) consisted of 3 parts of pure CHCl_3 and 1 part of distilled water. HCl produced by irradiation and the amount of HCl was found to be linear with dose. J. Tepliz and J. Bednar (71) have determined the radiolysis product HCl in the solution by titration with 0.001 N NaOH . The yield is independent of radiation intensity within a wide range and dosimeter can be used from 1000 to 40.000 rads. The resistance of irradiated aqueous solution of CHCl_3 was measured (72) and found that it is linear with dose in the range of $4 \cdot 10^3$ - $3 \cdot 10^5$ rads.

Chloroform and aqueous solution of indicator dye system has been suggested by G. V. Taplin and C. H. Douglas (73) for the range of 100 - 150 rads. The solution has a pH around 6.6-7.2 and contains 0.9-12 mg/cc of dye. Dithizone and methyl yellow each dissolved in CHCl_3 , CCl_4 and EtOH (74) have been investigated as a dosimeter for ionizing radiations. Acid formation from CHCl_3 or $\text{CHCl}_2\text{CHCl}_2$ in resorcinol is linear with doses greater than $2 \cdot 10^5$ rads (75).

Trichloroethylene saturated water and tetrachloroethylene systems offer good shelf life, dose rate independence, stability toward heat and light. With these systems it is possible to measure doses in the range of 5 to 10^6 rads (76).

Tetrachloroethylene system has been suggested by S. C. Sigoloff (77) to measure doses between $5 \cdot 10^5$ - $5 \cdot 10^6$ rads. The response was evaluated by using titration technique.

CCl_4 - EtOH dosimeter proposed by G. L. Clark and P. E. Biersteidt (74) and the system was improved by Z. Spurny (78) and adapted for use in the 0-200 rads range. HCl produced by irradiation and the pH of the aqueous phase was determined.

CCl_4 was used as chemical γ -dosimeter insensitive to neutrons. CCl_4 was not completely purified so that traces of CHCl_3 could remain as stabilizer. The color change of freshly prepared 10^{-4} M solution of

dithizone in CCl_4 (79) was found to be suitable for the spectrophotometric determination of doses from 5 to 600 rads at 610 nm. CCl_4 -dithizone system has been also studied by J. C. Pahissa et al. (80). The system shows absorbances at 615 nm and 400 nm. The sum of the absorbances is a useful parameter for dosimeter purposes. This parameter is independent of dose rate, photon energy and dithizone concentration. As a lower limit of operation of this dosimeter 20 rads can be measured with $\pm 10\%$ error. The upper limit depends on the dithizone concentration. Mixtures of CCl_4 and various alcohols were used as a γ -ray dosimeter system by K. Hannerz (81). Some of the mixtures were very sensitive to ionizing radiation but unstable and sensitive to impurities.

BENZENE, BENZOATE AND CHLOROBENZENE DOSIMETERS

The quantity of phenol formed in aerated aqueous solutions of benzene irradiated with or without addition of CdSO_4 , H_3BO_3 , LiBO_2 has been investigated by T. R. Johnson and J. J. Martin (82). The production of phenol from benzene was used to measure doses as low as 10 rads by L. I. Kartasheva and A. K. Pikaev (83). 2, 6-dibromo-N-chlorobenzoquinoneimine was used as indicator. The system is capable of measuring doses in the range of 20-35.000 rads.

Chromotographic determination of phenol suggested by B. Caire and L. Ordinaire (84) in the gaseous phase makes it possible to work with very small quantities of liquid. The system permits to measure doses between 10.000 and 70.000 rads range. M. E. J. Carr (85) has improved the method by measuring the phenol produced in dilute aqueous benzene solution by means of the increase in light absorption at 270 nm.

G. Stein and J. Weiss (86) have showed that the yields of benzene and benzoic acid depend on the pH of the solution. Benzene and sodium benzoate (87) were found to meet requirements of measuring the radiation doses with the formation of phenolic products which are readily estimated colorimetrically. Yields are not critically dependent on the pH, concentration and the temperature of the solutions. Yields are directly proportional to the doses up to $6 \cdot 10^4$ rads. The benzene dosimeter was used with Al, Ni, Cu and stainless steel containers (88). Within the precision of the method the Al, stainless steel and Ni gave identically good results.

Aqueous calcium benzoate system has been studied by W. A. Armstrong and D. W. Grant (89). They found that the system meets

many of the requirements for a sensitive chemical dosimeter. The concentration of salicylic acid determined spectrophotometrically increases linearly with radiation doses from 5 to 5.000 rads. The system is independent of temperature and energy. The salicylic acid yield in the aqueous calcium benzoate dosimeter has been determined spectrophotometrically by W. A. Armstrong et al. (90) and found a value which is below of the previously published values for Co-60 γ -rays.

Chlorobenzene water system has been used as a γ -ray dosimeter (91). The HCl produced determined directly by titration with 0.001 N NaOH. The amount of HCl varies linearly with dose from $4.5 \cdot 10^6$ to $1.3 \cdot 10^8$ rads and is independent of temperature and dose rate. The radiolytic HCl yield formed from the chlorobenzene-ethanol system measured as a function of dose by I. Dvornik (92). The yield was only slightly sensitive to impurities and HCl concentration was determined by titration method directly in the irradiated sample.

SODIUM FORMATE DOSIMETER

Sodium formate in aqueous solution is a suitable chemical dosimeter for ionizing radiations and it covers 1-80 Megarads dose range (93). The effect of oxygen initially dissolved in the solution is almost negligible and the reaction due to irradiation is linear with dose. The system is independent of temperature in the range used. The fast electron radiolysis of aqueous formate solution was studied by G. Juppe et al. (94). The yield has been determined by titration method and found a value approximately equal to 3 for the oxidizable compound in acid and alkaline solutions.

Method	Analysis	Dose range (rads)	Reference
Fricke	Optical absorption 305 nm	$4 \cdot 10^3 - 4 \cdot 10^4$	1 - 15, 22 - 29
Fricke with Fe-59	Radiometry	$40 - 4 \cdot 10^4$	16, 17
Fricke with benzene and benzoic acid	Colorimetry	$10 - 4 \cdot 10^4$	18, 19
Fricke with Copper	Optical absorption 305 nm	$4 \cdot 10^4 - 10^7$	20, 21
Ceric	Optical absorption 320 nm	$10^5 - 10^7$	30 - 39
Potassium bichromate	Optical absorption 350 nm	$10^4 - 10^7$	40 - 43
Titanium sulfate	Optical absorption 410 nm	$10^5 - 10^8$	48
Ammonium molybdate	Optical absorption 690 nm	$10^4 - 10^7$	53
Oxalic acid	Colorimetry or titration	$10^6 - 10^8$	55 - 61
Chloral hydrate	Titration or electrical conductivity	$50 - 3 \cdot 10^4$	62 - 69
Halogenated hydrocarbons	Titration or optical absorption	$10 - 10^6$	70 - 81
Benzene, benzoate and chlorobenzene	Colorimetry, chromatography or optical absorption	$10 - 4 \cdot 10^4$	82 - 92
Sodium formate	Titration	$10^6 - 8 \cdot 10^7$	93, 94

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