

## COMPARATIVE INVESTIGATION OF THE AQUEOUS TECHNETIUM-TRICARBONYL IN DIFFERENT CHROMATOGRAPHIC SYSTEMS

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Technetium-tricarbonyl is well known as one of the moiety for the preparation of the radiopharmaceutically acceptable compounds. Present paper deals with the comparative investigation of chromatographic behavior of  $[\text{Tc}(\text{CO})_3(\text{H}_2\text{O})_3]^+$  and some of its complexes, which were prepared in aqueous solutions.

To prepare an aqueous solution of technetium-tricarbonyl we used two methods.

The method described by R. Alberto et al. [1]: 4.0 mg  $\text{Na}_2\text{CO}_3$ , 5.5 mg  $\text{NaBH}_4$  and 20mg KNa-tartrate were put together into a 10-ml vial, which was closed and flushed for 10 min with CO. 3 ml of an eluate from  $^{99\text{m}}\text{Tc}$  generator was added and the solution was heated to 75 °C for 30 min. The authors recommended using for quality control the TLC on silica gel plates with methanol/conc.HCl (99:1) as a mobile phase. The  $R_f$  value is 0,4 for  $[\text{}^{99\text{m}}\text{Tc}(\text{CO})_3(\text{H}_2\text{O})_3]^+$  (I) and 1,0 for  $^{99\text{m}}\text{TcO}_4^-$ .

The second method was one-stage direct carbonilation of pertechnetate ions ( $^{99}\text{Tc}$  or  $^{99\text{m}}\text{Tc}$ ) with carbon monoxide gas in acidic aqueous solution (pH 0.5 - 1.0) at 160 - 180 °C under pressure (12 - 15 MPa) [2,3]. TLC on silica gel plates in acetonitrile/acetic acid (5:1) was recommended for quality control. The  $R_f$  value is 0.6 for I and 1.0 for  $^{99\text{m}}\text{TcO}_4^-$ .

$[\text{}^{99}\text{Tc}(\text{CO})_3(\text{H}_2\text{O})_3]^+$  was used as a standard. This standard was prepared by the hydrolysis of  $^{99}\text{Tc}(\text{CO})_5\text{Cl}$  with the following identification of  $[\text{}^{99}\text{Tc}(\text{CO})_3(\text{H}_2\text{O})_3]^+$  by NMR-spectroscopy.

To evaluate the radiochemical yield of the obtained complexes we used TLC on silica gel plates (Merck, 5554) in the following systems:

- acetonitrile : acetic acid - 5:1 (A)
- methanol : conc.HCl - 99:1 (B)
- 0.1 M acetic acid (C)

In each experiment the strip was cut after autoradiography and the activity of each piece was measured. The spot of  $[\text{}^{99}\text{Tc}(\text{CO})_3(\text{H}_2\text{O})_3]^+$  was also detected in ultraviolet light using silica gel plates with fluorescent indicator (F<sub>254</sub>).

After the chromatography of the standard solution in the system A we observed a small activity at the origin and blurring spot with  $R_f$  value 0.3 - 0.8 (see Fig., b). So we decided not to use this system.

On the chromatogram of the standard solution in the system B  $[\text{}^{99}\text{Tc}(\text{CO})_3(\text{H}_2\text{O})_3]^+$  has the  $R_f$  value 0.4, but were also observed a small adsorption of activity from the origin up to a spot (see Fig., e).

The best results we obtained in the system C. The single compact spot was observed on the strip with  $R_f$  value 0.4 - 0.45 (see Fig., c). If we used silica gel plates of another firms (Kavalier, Sorbfil) the  $R_f$  value may be 0.5 or 0.6 (see Fig., d, h).

The analysis of aqueous alkaline solutions of I (volume activity 37 - 370 MBq/ml) was carried out in two systems - B and C. In the system B we observed the formation of I with the yield of about 40 % and about 20 % of activity was remained at the origin and at the solvent front. The great amount of activity presented the adsorption zone ( 30 - 35 %). In the system C the yield of I was 70 %, the impurities at the origin were about 10 % and about 20 % of the activity was at the spot that was corresponded to  $^{99m}\text{TcO}_4^-$  (see Fig., g).

If we synthesized I in the presence of histidine ( $10^{-3}$  M), in the system C we observed 80 % of I, less than 5 % at the origin and not more than 10 % in the form of  $^{99m}\text{TcO}_4^-$  (see Fig., h).

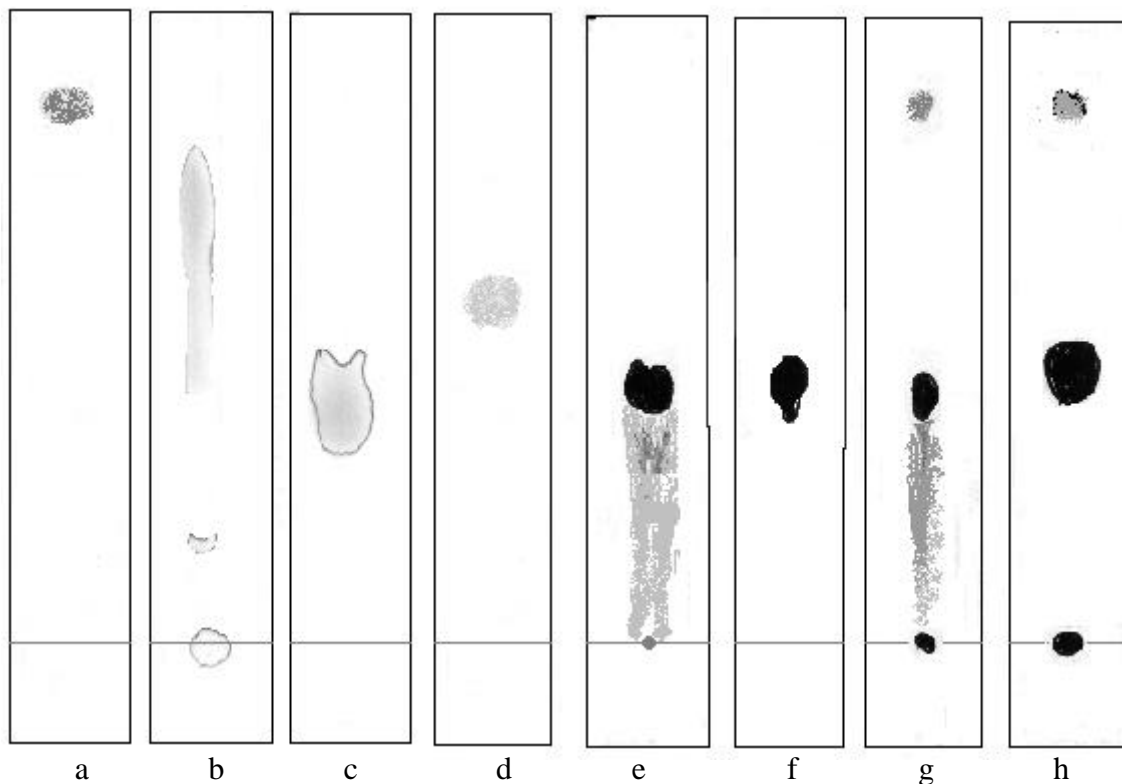


Fig. TLC chromatograms of  $^{99}\text{Tc}$  and  $^{99m}\text{Tc}$ -preparations.

- a)  $\text{K}^{99}\text{TcO}_4$ , silica gel plates, Merck 5554, in system A, B or C, detected in UV-light
- b)  $[\text{}^{99}\text{Tc}(\text{CO})_3(\text{H}_2\text{O})_3]^+$ , silica gel plates, Merck 5554, in system A, detected in UV-light
- c)  $[\text{}^{99}\text{Tc}(\text{CO})_3(\text{H}_2\text{O})_3]^+$ , silica gel plates, Merck 5554, in system C, detected in UV-light
- d)  $[\text{}^{99}\text{Tc}(\text{CO})_3(\text{H}_2\text{O})_3]^+$ , silica gel plates, Sorbfil, in system C, detected in UV-light
- e)  $[\text{}^{99}\text{Tc}(\text{CO})_3(\text{H}_2\text{O})_3]^+$ , silica gel plates, Merck 5554, in system B, autoradiography
- f)  $[\text{}^{99}\text{Tc}(\text{CO})_3(\text{H}_2\text{O})_3]^+$ , silica gel plates, Merck 5554, in system C, autoradiography
- g)  $[\text{}^{99m}\text{Tc}(\text{CO})_3(\text{H}_2\text{O})_3]^+$ , prepared using method [1], silica gel plates, Merck 5554, in system C, autoradiography
- h)  $[\text{}^{99m}\text{Tc}(\text{CO})_3(\text{H}_2\text{O})_3]^+$ , prepared using method [1] in the presence of histidine, silica gel plates, Kavalier, in system C, autoradiography

## CONCLUSION:

- for the correct identification of the reaction products by TLC the combination of two chromatographic systems should be used;
- in 0.1 M acetic acid practically no adsorption takes place;
- one can observe the formation of hydrolyzed form and  $\text{TcO}_4^-$  ions in the alkaline solutions of the  $^{99\text{m}}\text{Tc}$ -tricarbonyl.

## REFERENCES

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