4–7 Improving Adsorption Capacity for Medical Radioisotopes
—Elucidation of Molybdate Ion Adsorption Mechanism in Alumina—

Table 4-1 Mo adsorption capacity of alumina and its element content in milked solution
When the pH of the solution was lower, alumina adsorbed more molybdate ions, and Mo desorption and Al contamination also tended to increase.

pH of solution	Mo adsorption capacity (mg-Mo/g)	Contents in milked solution		
		Number of times	Mo desorption amount (mg-Mo/g-Al_2O_3)	Amount of Al (mg)
pH2	74.1	1st	2.77	0.08
		2nd	2.33	0.06
pH4	61.5	1st	0.88	0.02
		2nd	0.92	0.02
pH6	53.8	1st	0.68	–
		2nd	0.62	–

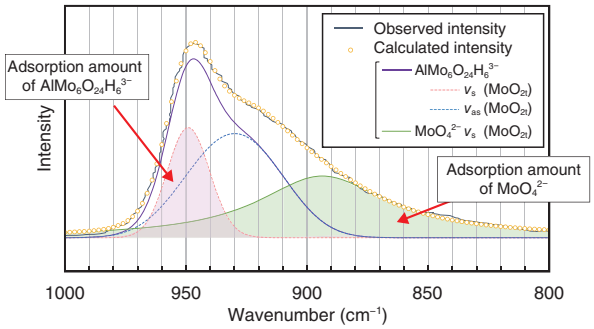

Fig.4-15 Adsorption of molybdate ions in the Raman spectrum (for pH 4)
The adsorption amounts of two molybdate ion species were compared considering their peak areas by fitting the Raman spectrum with the two ion peaks.

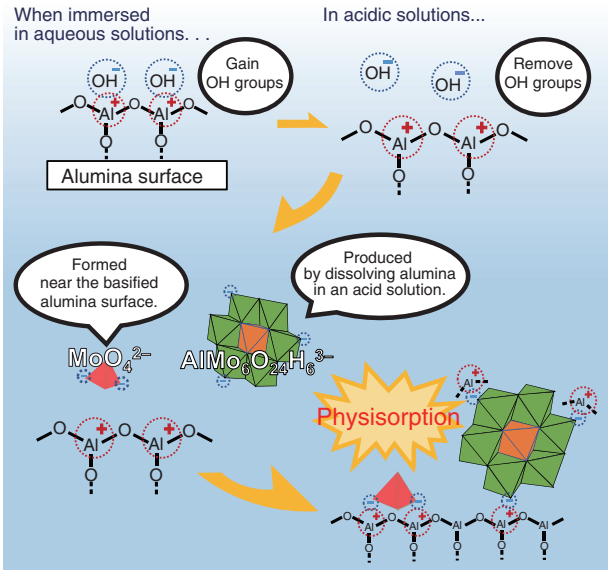

Fig.4-16 Mechanism of molybdate ion adsorption in alumina
i) Alumina gains OH groups on the surface when immersed in an aqueous solution. ii) When alumina is immersed in an acidic solution, the OH groups are removed, and the alumina surface becomes positively charged. iii) The release of OH^- makes the vicinity of the alumina surface basic and drives the formation of MoO_4^{2-}. iv) A small amount of alumina is dissolved in the acidic solution, and $AlMo_6O_{24}H_6^{3-}$ is produced. v) These two anions are physisorbed at the positively charged sites on the alumina surface.

Technetium-99m (^{99m}Tc), a representative nuclide for radiopharmaceutical, is a decay product of molybdenum-99 (^{99}Mo). At present, ^{99}Mo is produced by the fission of uranium (i.e., the fission method), but the activation method is attracting attention from the viewpoint of nuclear security. This method produces ^{99}Mo via the neutron irradiation of ^{98}Mo. However, compared to the fission method, the activation method can produce only a small amount of ^{99}Mo. Extraction of ^{99m}Tc from ^{99}Mo is based on the phenomenon that only ^{99m}Tc is eluted upon passing saline through alumina (Al_2O_3) adsorbed ^{99}Mo (milking). To extract an amount of ^{99m}Tc equivalent to that extracted by the fission method, it is necessary to improve the Mo adsorption capacity of alumina. In this work, we focused on the molybdate ion, which is the Mo form adsorbed on alumina, and elucidated the adsorption mechanism.

First, alumina was immersed in solutions of different pH containing molybdate ions and was milked by a saline solution. More Mo was adsorbed at lower pH values, but the milked solution contained more Mo and Al (Table 4-1). Next, the amount of OH groups on the alumina surface increased at pH 4 and pH 6 compared to that before adsorption, but almost no change was observed at pH 2. Therefore, the OH groups on the surface increased upon immersion in the solution, but the formation of OH groups was suppressed when a large amount of Mo was adsorbed. Furthermore, the adsorbed molybdate ion species were investigated and were found to be ions containing Mo(VI). Finally, the adsorbed molybdate ion species were identified as MoO_4^{2-} and $AlMo_6O_{24}H_6^{3-}$, and these adsorption amounts were compared (Fig.4-15). From the increasing content of Mo and Al in milked solution with lower pH and the adsorption amount of two molybdate ion species, it was estimated that $AlMo_6O_{24}H_6^{3-}$ has weak adsorbability.

Based on this result, we elucidated the Mo adsorption mechanism in alumina (Fig.4-16). The approaches found to improve the Mo adsorption capacity are as follows: 1) increasing the amount of surface OH groups by enlarging the surface area or controlling the crystal phase, and 2) optimizing the Mo solution for adsorption as MoO_4^{2-} ions. The results of this work will contribute to improve the adsorption property of alumina and the practical application of the activation method with excellent nuclear security.

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Reference

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