

HIGH-PURITY TUNGSTEN(VI) OXYTETRACHLORIDE AND PROCESS FOR PREPARING SAME

The present invention relates to tungsten(VI) oxytetrachloride which is notable for particular chemical purity and phase purity, and also to a method for its production.

Tungsten(VI) oxytetrachloride is an inorganic compound of tungsten from the group of the oxychlorides, having the stoichiometric formula WOCl_4 . Tungsten(VI) oxytetrachloride is used primarily as a catalyst for the production of olefins, especially in ring-opening reactions. Further fields of use for tungsten(VI) oxytetrachloride are found in the semiconductor industry and as a starting compound in sol-gel processes.

DE 33 47 918 describes a glass substrate having a thermochromic coating containing VO_2 or V_2O_3 , with variable transmissibility for solar rays, where an optical change takes place within a temperature range from 25 to 75°C and the transmissibility for the entire solar energy in the infrared range decreases by a factor of at least two when the coating is heated through its temperature transition range. The coating containing VO_2 or V_2O_3 is a layer which is produced by chemical decomposition and optionally reduction of a chemically deposited vanadium compound and which is doped with oxides of metals having a larger ionic radius than vanadium. Among suitable dopants identified is WOCl_4 , which when added is able to lower the transformation temperature of the VO_2 film.

N. Özer et al., in their essay *"Optical and Electrochromic Properties of Sol-Gel Deposited Doped Tungsten Oxide Films"* from January 1996, describe the use of WOCl_4 as a precursor compound in the production of films doped with tungsten oxide.

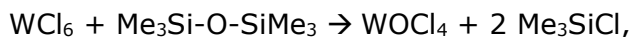
The skilled person knows of various methods for the production of tungsten(VI) oxytetrachloride, as for example by reaction of tungsten(VI) oxide, tungsten(VI) chloride or sodium tungstate with thionyl chloride, or by thermal decomposition of tungsten(VI) dioxide dichloride. Other production methods include the boiling of tungsten(VI) oxide in octachlorocyclopentene or the reaction of stoichiometric amounts of tungsten(VI) oxide with tungsten(VI) chloride in an evacuated

ampoule at 200°C.

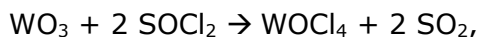
F. Zado, in his essay *"The high yield synthesis of the tungsten(VI) oxyhalides WOCl₄, WOBr₄ and WO₂Cl₂ and some observations on tungsten(VI) bromide and tungsten(V) chloride"*, published in *J. Inorg. Nucl. Chem.* 25, 1115 (1963),
5 describes the preparation of tungsten(VI) oxytetrachloride by sealing WCl₆ and WO₃ in an evacuated ampoule and heating the ampoule to 100°C. The temperature was raised after two hours to 150°C and the ampoule was positioned so that the deposited product could be sublimed over a period of 24 hours.

10 A.E. Castro Lunar et al., in *J. Chem. Eng. Data* 1983, 26, 349-350, under the title *"Vapour Pressure of WOCl₄"*, describe the preparation of tungsten(VI) oxytetrachloride by reaction of CCl₄ with WO₃ at 673 K.

Examples of other known preparation methods for tungsten(VI) oxytetrachloride are the reaction of WCl₆ with an organosilicon compound according to the
15 equation



the aforementioned use of thionyl chloride according to the equation



and also the one-step reaction of heated tungsten in oxygen-containing
20 Cl₂ gas.

The methods described in the prior art have the disadvantage, however, that the tungsten(VI) oxytetrachloride usually has a significant fraction of impurities, such as metallic impurities and other tungsten compounds, which are manifested in further crystal phases, and the methods can be employed only on a laboratory
25 scale. Hence the chemical purity of commercially available tungsten(VI) oxytetrachloride is stated at 98% to 99.95%, based on metallic impurities. The majority of the metallic impurities can certainly be removed by further purification steps, such as sublimation. The sublimation, however, results in increased crystal growth, and so the tungsten(VI) oxytetrachloride takes the
30 form of large, acicular crystals having a low bulk density, which for a series of

applications are a disadvantage. To obtain suitable particle sizes, the crystals have to be processed appropriately by grinding methods, for example. Owing to the low stability of tungsten(VI) oxytetrachloride, however, this leads to the introduction of metallic impurities and to the formation of unwanted phases. A
5 further, exacerbating factor is that the decomposition of tungsten(VI) oxytetrachloride is accompanied by the formation of highly corrosive compounds such as hydrochloric acid. The purification methods that are conventionally required also have the disadvantage that some impurities, especially further tungsten compounds and certain non-metals, cannot be removed sufficiently, if
10 at all.

It is therefore an object of the present invention to provide tungsten(VI) oxytetrachloride that overcomes the disadvantages of the prior art, and also a simple and efficient method for its production, which can be implemented even on an industrial scale and which operates without additional steps of mechanical
15 work-up, such as grinding, sieving or subliming, for example.

This object is achieved through the provision of tungsten(VI) oxytetrachloride having a high chemical and crystallographic purity and a specific morphology.

A first subject of the present application is therefore a tungsten(VI) oxytetrachloride which has a chemical purity of greater than 99.95% and where
20 the tungsten(VI) oxytetrachloride has a fraction of compounds selected from the group consisting of WCl_6 , WO_2Cl_2 , WO_3 and WO_2 , defined as the ratio of the reflection having the highest intensity of one of these compounds ($I(P2)100$) in the x-ray diffraction pattern to the reflection having the highest intensity of the tungsten(VI) oxytetrachloride ($I(WOCl_4)100$) in the x-ray diffraction pattern,
25 expressed as $I(P2)100/I(WOCl_4)100$, of less than 0.03.

The tungsten(VI) oxytetrachloride in accordance with the present invention is more particularly tungsten(VI) oxytetrachloride of the empirical formula $WOCl_4$.

In the context of the present invention, the chemical purity of a $WOCl_4$ sample refers to the ratio of the accumulated weight proportions of the element's
30 tungsten, chlorine and oxygen to the accumulated weight proportions of all other impurifying elements in the sample. The purity of the substance is expressed as a quantitative ratio of $WOCl_4$ to the overall weight of composition, with a

substance having a chemical purity of 100% if there are no other elements present except tungsten, chlorine and oxygen. Impurities in the sense of the present invention encompass not only metallic impurities but also non-metallic impurities, in particular silicon, carbon, sulphur, molybdenum and nickel and components thereof.

The chemical purity has to be distinguished from the crystallographic purity. In the context of the present application, the crystallographic purity of a substance is the ratio of the crystal phase of an unwanted compound to the crystal phase of the desired compound, presently tungsten(VI) oxytetrachloride. The ratio of the crystal phases here can be determined, for example, by means of the intensities of the various phases in an x-ray diffraction pattern. Suitable recordings can be made on powder samples using, for example, an instrument from Malvern-PANalytical (X'Pert-MPD with semiconductor detector, Cu LFF x-ray tube with 40 KV/40 mA, Ni filter).

Besides the stated chemical impurities, conventional tungsten(VI) oxytetrachloride may comprise further compounds, such as, for example, WCl_6 , and optionally further tungsten oxides as what are called secondary phases. These oxides are, primarily, the related oxides WO_2Cl_2 , WO_3 and WO_2 , which possess a different crystal structure from tungsten(VI) oxytetrachloride and are detectable via x-ray diffractometry. In one particularly preferred embodiment, the tungsten(VI) oxytetrachloride of the invention comprises substantially no further tungsten compounds and in particular no further tungsten oxide compounds. Preferably the tungsten(VI) oxytetrachloride of the invention is substantially free of compounds selected from the group consisting of WCl_6 , WO_2Cl_2 , WO_3 and WO_2 . The fraction of further compounds as recited above may be defined by way of the intensity ratio of the main tungsten(VI) oxytetrachloride phase to the stated secondary phases. The main and secondary phases may be ascertained by means of their reflection intensities in the x-ray diffraction pattern, indicated there as impulses per angle $[\circ 2\theta]$. In one particularly preferred embodiment, the ratio of the reflection having the highest intensity of a secondary phase ($I(\text{P}2)_{100}$) to the reflection having the highest intensity of the main tungsten(VI) oxytetrachloride phase ($I(\text{WOCl}_4)_{100}$), expressed as $I(\text{P}2)_{100}/I(\text{WOCl}_4)_{100}$, is preferably less than 0.02, more preferably 0.001 to 0.02, more particularly 0.001 to 0.01, determined in each

case by means of x-ray diffractometry. The tungsten(VI) oxytetrachloride of the invention is regarded in the context of the present invention as being substantially free of compounds selected from the group consisting of WCl_6 , WO_2Cl_2 , WO_3 and WO_2 and declared as secondary phases in the context of the present invention if the ratio of the main and secondary phases as defined above is 0.001 or less. In a further preferred embodiment, no secondary phases are detectable. The fractions of further tungsten compounds are ascertained here by means of x-ray diffractometry, determined for example on powder samples using an instrument from Malvern-PANalytical (X'Pert-MPD with semiconductor detector, Cu LFF x-ray tube with 40 KV/40 mA, Ni filter).

In one preferred embodiment the tungsten(VI) oxytetrachloride of the invention has a chemical purity of 99.99% or more, preferably 99.995% or more and more preferably 99.999% or more.

Conventional tungsten(VI) oxytetrachloride generally has a series of chemical impurities which can have a disruptive influence on further uses of the tungsten(VI) oxytetrachloride. These chemical impurities, depending on production method, are, more particularly, silicon and its compounds, sulfur and its compounds, and also carbon, molybdenum and iron. In contrast to this, the tungsten(VI) oxytetrachloride of the invention is notable for a low level of impurities. In one preferred embodiment the tungsten(VI) oxytetrachloride of the invention comprises silicon and/or compounds of silicon in an amount of less than 50 ppm, preferably in an amount of less than 25 ppm and more preferably in an amount of less than 5 ppm, the data being based in each case on the total weight of the tungsten(VI) oxytetrachloride.

In a further preferred embodiment, the tungsten(VI) oxytetrachloride of the invention comprises sulfur and/or compounds of sulfur in an amount of less than 100 ppm, preferably in an amount of less than 30 ppm and more preferably in an amount of less than 10 ppm, the data being based in each case on the total weight of the tungsten(VI) oxytetrachloride.

The fraction of metallic impurities, apart from silicon, overall is less than 100 ppm, preferably less than 70 ppm and more preferably less than 50 ppm, with the data being based in each case on the total weight of the tungsten(VI)

oxytetrachloride. The metallic impurities may be, for example, elements from main groups 2 and 3, refractory metals, and more particularly iron, chromium, nickel and molybdenum. In one particularly preferred embodiment, the tungsten(VI) oxytetrachloride of the invention has a fraction of molybdenum and/or its compounds of less than 20 ppm, preferably less than 10 ppm and more preferably less than 5 ppm, based in each case on the total weight of the tungsten(VI) oxytetrachloride.

Likewise preferred is an embodiment in which the tungsten(VI) oxytetrachloride of the invention comprises carbon in an amount of less than 200 ppm, preferably in an amount of less than 100 ppm and more preferably in an amount of less than 40 ppm, the data being based in each case on the total weight of the tungsten(VI) oxytetrachloride.

As well as its high chemical and crystallographic purity, moreover, the tungsten(VI) oxytetrachloride of the invention is notable for its morphology. Whereas conventional tungsten(VI) oxytetrachloride generally takes the form of large acicular crystals, this has surprisingly not been observed with the tungsten(VI) oxytetrachloride of the invention. In one preferred embodiment, therefore, the tungsten(VI) oxytetrachloride of the invention takes the form of a fine powder, with 90% of all the powder particles having a particle size of 100 μm or less, preferably of 70 μm or less, determined by means of optical microscopy, and where the particle size denotes the longest extent of the particle. The percentage figure here is based on the total number of powder particles. It has surprisingly been found that as a result of the specific morphology of the tungsten(VI) oxytetrachloride of the invention, it has been possible to improve the dissolution kinetics of the tungsten(VI) oxytetrachloride in the further application.

As a result of the specific morphology of the tungsten(VI) oxytetrachloride of the invention, moreover, it has been possible to achieve a distinct improvement in the bulk density, sometimes also referred to as bulk weight. In one preferred embodiment the tungsten(VI) oxytetrachloride of the invention has a bulk density of more than 0.5 g/cm³. The bulk density ρ_b is defined as the ratio of the mass m of the bed to the bed volume occupied, V_b , according to the formula

$\rho_b = \frac{m}{V_b}$ and can be determined by means of ASTM B329-06.

A further subject of the present invention is a method for producing the tungsten(VI) oxytetrachloride of the invention. The method of the invention is notable for the fact that in a first step a) tungsten metal is reacted with chlorine gas and oxygen at a temperature T(1) and the resulting product mixture is subjected in a further step b) to an oxidation at a temperature T(2) in the presence of an oxidizing agent to give the tungsten(VI) oxytetrachloride of the invention.

The tungsten metal used in step a) is preferably a metal powder having a particle size D90 of less than 300 μm , preferably having a particle size D90 of less than 150 μm , determined by means of ASTM B822. The D90 of the particle size here denotes the fraction of particles which have a particle size less than the specified value.

The reaction in step a) of the method of the invention is conducted at a temperature T(1) of 600 to 1000 $^{\circ}\text{C}$, preferably.

Without being tied to a particular theory, it is assumed in the context of the present invention that in step a) of the method of the invention, the reaction of the tungsten metal with chlorine gas and oxygen produces a mixture of tungsten(VI) oxytetrachloride and tungsten(VI) chloride. In the context of the method of the invention it has surprisingly been found that in a further step, the tungsten(VI) chloride obtained can be selectively oxidized to tungsten(VI) oxytetrachloride, giving tungsten(VI) oxytetrachloride having a high purity and a specific morphology.

The method of the invention affords the advantage, moreover, that it furnished tungsten(VI) oxytetrachloride in high chemical and crystallographic purity and so makes it unnecessary to employ further work-up steps such as subliming, for example. Preferred, therefore, is an embodiment of the method of the invention in which further purification steps and/or operating steps, especially subliming and/or grinding, are omitted. Without being tied to a particular theory, it is assumed that the omission of additional operating steps, such as subliming, grinding or sieving, contributes to the production, by virtue of the method of the invention, of tungsten(VI) oxytetrachloride which is present in the form of small

particles, in contrast to conventional tungsten(VI) oxytetrachloride.

The oxidizing agent used in step b) is preferably selected from the group consisting of oxygen, water and steam. A particularly preferred oxidizing agent used in step b) of the method of the invention is water or steam. When water is
5 used as oxidizing agent in step b) of the method of the invention, the oxidation is preferably carried out at a relative atmospheric humidity of 20% to 80%, more preferably of 40% to 60%.

The oxidation of tungsten chloride compounds takes place conventionally at temperatures of 300°C or more. In the context of the method of the invention
10 it has surprisingly been found that the oxidation may also be performed at significantly lower temperatures without the formation of higher oxides or oxychlorides, such as WO_2Cl_2 or WO_3 , for example, and without any detractions from the yield, especially when using water as oxidizing agent. Preferred, therefore, is an embodiment of the method wherein the oxidation in step b) is
15 performed at a temperature $T(2)$ of 0 to 80°C, preferably 0 to 60°C, preferably for a period of 5 minutes to 50 hours.

It has proven to be advantageous, furthermore, to perform the reaction of the tungsten metal with oxygen and chlorine gas, contrary to the general expectation, in an atmosphere in which the oxygen is in a substoichiometric ratio
20 in relation to the tungsten metal. Preferred, therefore, is an embodiment wherein the oxygen in step a) is used in a substoichiometric ratio. The stoichiometric ratio of oxygen to tungsten metal in step a) of the method of the invention is preferably 0.85 to 0.97, more preferably 0.85 to 0.9.

A series of methods known in the prior art for producing tungsten(VI)
25 oxytetrachloride are based on the use of organic compounds, especially organic sulfur compounds and organosilicon compounds. In the context of the present invention it has surprisingly been found that tungsten(VI) oxytetrachloride can be obtained in high purity under mild conditions even without the use of such compounds, some of which are injurious to health. Preferred, therefore, is an
30 embodiment of the method of the invention in which no sulfur compounds and/or silicon compounds and/or chlorinated hydrocarbons are used in the method of the invention.

The tungsten(VI) oxytetrachloride produced according to the method of the invention differs not only in terms of its purity but also, in particular, in terms of its morphology from tungsten(VI) oxytetrachloride produced by means of conventional methods. A further subject of the present invention, therefore, is tungsten(VI) oxytetrachloride which has been produced according to the method of the invention. A feature of the tungsten(VI) oxytetrachloride produced by means of the method of the invention is that 90% of all the particles have a particle size of 100 μm or less, preferably 70 μm or less, determined by means of optical microscopy. The percentages here are based on the total number of the particles. The particle size here denotes the longest dimension of the particle.

A further subject of the present invention is the use of the tungsten(VI) oxytetrachloride of the invention or of the tungsten(VI) oxytetrachloride obtained according to the method of the invention as a catalyst in chemical reactions, more particularly in the preparation of functional hydrocarbons; in the semiconductor industry, and/or in sol-gel processes, especially for producing electrochromic coatings.

The present invention is elucidated in more detail with the following examples and figures, which should not be understood in any way as limiting the concept of the invention.

Examples:

1. Comparative example 1

Tungsten(VI) oxytetrachloride was prepared according to the following reaction equation:



An analysis of the tungsten(VI) oxytetrachloride obtained, including the chemical purity, the crystalline phases detected, the particle size and the bulk density, is collated in Table 1.

10 As is apparent from Figure 1, which shows an x-ray diffraction pattern of the tungsten(VI) oxytetrachloride obtained, there were significant fractions of WO_2Cl_2 as well as the desired compound.

15 Figures 2a and 2b show optical micrographs of the tungsten(VI) oxytetrachloride prepared according to the formula Comp. 1, with the acicular shape of the crystals being clearly apparent and less than 40% of all the particles having a size of less than 100 μm .

2. Comparative example 2

As a further comparison, tungsten(VI) oxytetrachloride was prepared according to the following reaction equation:



An analysis of the tungsten(VI) oxytetrachloride obtained, including the chemical purity, the crystalline phases detected, the particle size and the bulk density, is collated in Table 1.

25 An x-ray diffraction pattern of the product demonstrates the presence of WO_2Cl_2 as a secondary product, as well as the desired tungsten(VI) oxytetrachloride (Figure 3).

Figure 4 shows an optical micrograph of the tungsten(VI) oxytetrachloride

prepared according to the formula Comp. 2, with the acicular shape of the crystals being clearly apparent and less than 10% of all the particles having a size of less than 100 µm.

5 3. Comparative example 3

As a further comparison, tungsten(VI) oxytetrachloride was prepared according to the following reaction equation in a one-step reaction at 800°C:



10 An analysis of the tungsten(VI) oxytetrachloride obtained, including the chemical purity, the crystalline phases detected, the particle size and the bulk density, is collated in Table 1.

15 The tungsten(VI) oxytetrachloride obtained gave an x-ray diffraction pattern (Figure 5) which as well as the desired tungsten(VI) oxytetrachloride also contained significant amounts of WCl_6 and also WO_2Cl_2 . It should be noted that in this experiment, indeed, WO_2Cl_2 constitutes the main phase, and tungsten(VI) oxytetrachloride only a secondary phase. Reproduction tests under identical conditions confirmed the incidence of these three compounds in changing proportions, based on x-ray diffraction analyses. It was not possible to produce a pure-phase tungsten(VI) oxytetrachloride by the one-step reaction.

20 Optical micrographs of the tungsten(VI) oxytetrachloride prepared according to the formula Comp. 3 also clearly showed an acicular form of the crystals, with less than 50% of all the particles having a size of less than 100 µm.

Experi- ment	X-ray diffractometry		Particle size (optical microscopy)	Chemical trace analysis										
	Qualitative phase composition	Quantitative phase composition I(P2)100 / I(WOCl4)100		Si	S	C	Mo	Fe	Sum total of all metallic impurities	Sum total of all impurities including all non-metals and Si	Chemical purity	Bulk density		
	MP Main phase SP Secondary phases		[%]	[ppm]	[ppm]	[ppm]	[ppm]	[ppm]			[ppm / wt%]	[%]	[g/cm3]	
Comp. 1	WOCl ₄ (MP), WO ₂ Cl ₂ (SP)	0.052	<40%	55	21	981	10	71	135	1192 / 0.1192	99.8808	0.43		
Comp. 2	WOCl ₄ (MP), WO ₂ Cl ₂ (SP)	0.108	<10%	15	352	240	11	80	104	711 / 0.0711	99.9289	0.33		
Comp. 3	WO ₂ Cl ₂ (MP)WOCl ₄ (SP), WCl ₆ (SP)	1.19	<50%	6	11	21	9	10	68	106 / 0.0106	99.9894	0.39		

Table 1

4. Inventive example

5 Tungsten(VI) oxytetrachloride was prepared in accordance with the invention by a two-step process, whereas in a first step a) tungsten metal powder having a particle size D90 of less than 150 μm was brought to reaction at 800 $^{\circ}\text{C}$ in a Cl_2 gas stream and with an addition of oxygen in a stoichiometric ratio of 0.85, based on the amount of tungsten used.

10 The received intermediate reaction product contains, due to the substoichiometric addition of oxygen (stoichiometric ratio of 0.85) beside the desired tungsten(VI) oxytetrachloride significant amounts of WCl_6 .

15 This intermediate was reacted according to step b) under various oxidation conditions to give the tungsten(VI) oxytetrachloride of the invention; the oxidation conditions and also all relevant analytical results for the powders produced under experiments 4.1 to 4.8 are collated in Table 2. The oxidation took place in a conditioning cabinet with water, with the cabinet controlling the relative atmospheric humidity and the temperature as indicated in Table 2.

20 An analysis of the tungsten(VI) oxytetrachloride obtained, including the chemical purity, the crystalline phases detected, the particle size and the bulk density, is evident from Table 2 under experiment 4.0.

Experiment	Reaction conditions (step b)			Results		Particle size (optical microscopy)	Chemical trace analysis						Sum total of all metallic impurities [ppm]	Sum total of all impurities including all non-metals and Si [ppm]	Chemical purity [%]	Bulk density [g/cm ³]
	Relative atmos. humidity	Temperature	Time	Qualitative phase composition	Quantitative phase composition I(P2)100 / I(WOCl4)100		Si	S	C	Mo	Fe					
	[%]	[°C]	[h]	MP main phase SP secondary phases		[%]	[ppm]	[ppm]	[ppm]	[ppm]	[ppm]		[ppm]	[ppm]	[%]	[g/cm ³]
4.0	Starting material from step a)			WOCl4(MP), WCl6(SP)	0.442	>95%	4	9	15	9	5		65	93	99.9907	0.59
4.1	60	5	1	WOCl4(MP), WCl6(SP)	0.01	>95%	3	9	16	8	8		60	88	99.9912	0.61
4.2	60	25	0.5	WOCl4	No sec. phase	>95%	4	9	12	9	6		63	88	99.9912	0.58
4.3	60	25	1	WOCl4	No sec. phase	>95%	4	8	17	7	9		67	96	99.9904	0.58
4.4	40	60	0.5	WOCl4	No sec. phase	>95%	2	9	16	9	6		65	92	99.9908	0.61
4.5	60	60	0.5	WOCl4	No sec. phase	>95%	4	6	19	6	7		68	97	99.9903	0.58
4.6	80	60	0.5	WOCl4(MP), WO2Cl2(SP)	0.0025	>95%	4	7	11	9	6		61	83	99.9912	0.63
4.7	60	90	0.2	WOCl4(MP), WO2Cl2(SP)	0.097	>95%	4	9	15	5	4		68	96	99.9904	0.65
4.8	60	90	0.5	WOCl4(MP), WO2Cl2(SP)	0.471	>95%	3	5	15	8	5		65	88	99.9912	0.64

Table 2

Figure 6 shows an x-ray diffraction pattern of the intermediate from experiment 4.0, obtained after step a). As is apparent from the pattern, there are no tungsten compounds other than the stated compounds WOCl_4 and WCl_6 .

5 Figure 7 shows an optical micrograph of the product after step a). In contrast to the conventional production methods, the powder is not in the form of acicular crystals. Instead, the particle size of more than 95% of the total number of the powder particles is less than 100 μm , based on the longest longitudinal dimension of the particles.

10 Figure 8 shows an x-ray diffraction pattern of the product from experiment 4.2, obtained after step b). As is apparent from the pattern, there was complete oxidation of the WCl_6 to give the desired WOCl_4 .

As shown by the optical micrograph in Figure 9, the morphology of the tungsten(VI) oxytetrachloride was not affected in any way by the oxidation. In the end product from experiment 4.2 as well, the tungsten(VI) oxytetrachloride
15 was in the form of small particles having a particle size of less than 100 μm for more than 95% of the overall number of the powder particles.

As is apparent from Table 2, in experiments 4.1 to 4.6, tungsten(VI) oxytetrachlorides of the invention were prepared that had a chemical purity of greater than 99.95% and contained no detectable secondary phases or secondary
20 phases with an $\text{I(P2)100/I(WOCl}_4\text{)100}$ ratio of less than 0.03, as a result, in particular, of control over the oxidation conditions during step b). If the process window for the oxidation is departed from, as in experiments 4.7 and 4.8, rapidly increasing amounts of WO_2Cl_2 are formed, becoming visible as a result of an increase in the $\text{I(P2)100/I(WOCl}_4\text{)100}$ ratio.

25 The fractions of crystalline tungsten compounds are ascertained by means of x-ray diffractometry, determined for example on powder samples using an instrument from Malvern-PANalytical (X'Pert-MPD with semiconductor detector, Cu LFF x-ray tube with 40 KV/40 mA, Ni filter). In the qualitative analysis of the x-ray diffraction patterns, the reflections ascertained were assigned to tungsten
30 compound reflections known from the literature.

The trace analysis of the chemical impurities was carried out by means of ICP-OES

using the analytical instruments PQ 9000 (Analytik Jena) or Ultima 2 (Horiba). The chemical purity ascertained is obtained arithmetically as the difference between the hypothetical contamination-free sample at 100 wt% and the sum total of all impurities, including all non-metals and Si, in wt%.

Claims:

1. Tungsten(VI) oxytetrachloride, characterized in that tungsten(VI) oxytetrachloride has a chemical purity of greater than 99.95% and where the tungsten(VI) oxytetrachloride has a fraction of compounds selected from the group consisting of WCl_6 , WO_2Cl_2 , WO_3 and WO_2 , defined as the ratio of the reflection having the highest intensity of one of these compounds ($I(\text{P}2)100$) in the x-ray diffraction pattern to the reflection having the highest intensity of the tungsten(VI) oxytetrachloride ($I(\text{WOCl}_4)100$) in the x-ray diffraction pattern, expressed as $I(\text{P}2)100/I(\text{WOCl}_4)100$, of less than 0.03.
2. Tungsten(VI) oxytetrachloride according to Claim 1, characterized in that the tungsten(VI) oxytetrachloride has a fraction of secondary phases, defined as the ratio of the reflection having the highest intensity of a secondary phase ($I(\text{P}2)100$) to the reflection having the highest intensity of the tungsten(VI) oxytetrachloride ($I(\text{WOCl}_4)100$), expressed as $I(\text{P}2)100/I(\text{WOCl}_4)100$, of less than 0.02, preferably 0.001 to 0.02, more preferably 0.001 to 0.01, in each case by means of x-ray diffractometry.
3. Tungsten(VI) oxytetrachloride according to at least one of Claims 1 and 2, characterized in that the tungsten(VI) oxytetrachloride has a chemical purity of 99.99% or more, preferably 99.995% or more, more preferably 99.999% or more.
4. Tungsten(VI) oxytetrachloride according to at least one of Claims 1 to 3, characterized in that the tungsten(VI) oxytetrachloride comprises silicon and/or compounds of silicon in an amount of less than 50 ppm, preferably less than 25 ppm, more preferably of less than 5 ppm, based in each case on the total weight of the tungsten(VI) oxytetrachloride.
5. Tungsten(VI) oxytetrachloride according to at least one of Claims 1 to 4, characterized in that the tungsten(VI) oxytetrachloride comprises sulfur and/or compounds of sulfur in an amount of less than 100 ppm, preferably less than 30 ppm, more preferably of less than 10 ppm, based in each case on the total weight of the tungsten(VI) oxytetrachloride.

6. Tungsten(VI) oxytetrachloride according to at least one of Claims 1 to 5, characterized in that the tungsten(VI) oxytetrachloride has a fraction of molybdenum and/or its compounds of less than 20 ppm, preferably less than 10 ppm, more preferably of less than 5 ppm, based in each case on the total weight of the tungsten(VI) oxytetrachloride.
7. Tungsten(VI) oxytetrachloride according to at least one of Claims 1 to 6, characterized in that the fraction of metallic impurities in the tungsten(VI) oxytetrachloride is less than 100 ppm, preferably less than 70 ppm, based in each case on the total weight of the tungsten(VI) oxytetrachloride.
8. Tungsten(VI) oxytetrachloride according to at least one of Claims 1 to 7, characterized in that the tungsten(VI) oxytetrachloride comprises carbon in an amount of less than 200 ppm, preferably less than 100 ppm, more preferably of less than 40 ppm, based in each case on the total weight of the tungsten(VI) oxytetrachloride.
9. Tungsten(VI) oxytetrachloride according to at least one of Claims 1 to 8, characterized in that the tungsten(VI) oxytetrachloride is in the form of a powder where 90% of all the powder particles have a particle size of 100 μm or less, preferably of 70 μm or less, determined by means of optical microscopy.
10. Tungsten(VI) oxytetrachloride according to at least one of Claims 1 to 9, characterized in that the tungsten(VI) oxytetrachloride has a bulk density of greater than 0.5 g/cm³.
11. Method for producing tungsten(VI) oxytetrachloride according to at least one of Claims 1 to 10, characterized in that in a first step a) tungsten metal is reacted with chlorine gas and oxygen and the resulting product mixture is subjected in a further step b) to an oxidation in the presence of an oxidizing agent to give tungsten(VI) oxytetrachloride.
12. Method according to Claim 11, characterized in that the oxidizing agent in step b) is selected from the group consisting of oxygen, water and steam.
13. Method according to at least one of Claims 11 and 12, characterized in that the oxidation in step b) is carried out at a relative atmospheric humidity of 20% to 80%, preferably 40% to 60% with water as oxidizing agent.

14. Method according to at least one of Claims 11 to 13, characterized in that the oxidation in step b) is performed at a temperature of 0 to 80°C, preferably 0 to 60°C.
15. Method according to at least one of Claims 11 to 14, characterized in that the oxygen in step a) is used in a substoichiometric proportion, with the stoichiometric ratio of oxygen to tungsten metal being preferably 0.8 to 0.97 and more preferably 0.85 to 0.9.
16. Tungsten(VI) oxytetrachloride obtainable by a method according to at least one of Claims 11 to 15.
17. Use of tungsten(VI) oxytetrachloride according to at least one of Claims 1 to 10 or 16 as a catalyst in chemical reactions, more particularly in the preparation of functional hydrocarbons; in the semiconductor industry, and/or in sol-gel processes, especially for producing electrochromic coatings.