REFRAM

Final conference: WP5
Substitution potential of refractory metals

9-10/3/2017

Funded by the Horizon 2020 Framework programme of the European Union (Grant Agreement Number 688993)

Mo, Re, Nb, Ta, W: VTT (Ta), IMN (Mo, Re), ICCRAM (W), TUDELFT (Nb)
Päivi Kivikytö-Reponen, Marjaana Karhu, Katarzyna Bilewska, Santiago Cuesta-Lopez, Rocío Barros, Erik Offerman

CONTENTS

- MSP-REFRAM project goals for substitution work package – general (10 min)
- Element specific (40 min)
  - Mo
  - Re
  - Nb
  - Ta
  - W – Dr. Santiago Cuesta-López
- Discussion and questions

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Substitution approaches

- Substitutability assessment – methodology to evaluate, indexing
  - EU study of CRMs
  - Scaling 0.0-1.0
- Substitutes development – development of substitutes - materials science approach – application approach
  - scientific literature review of development
  - patents
  - hidden development

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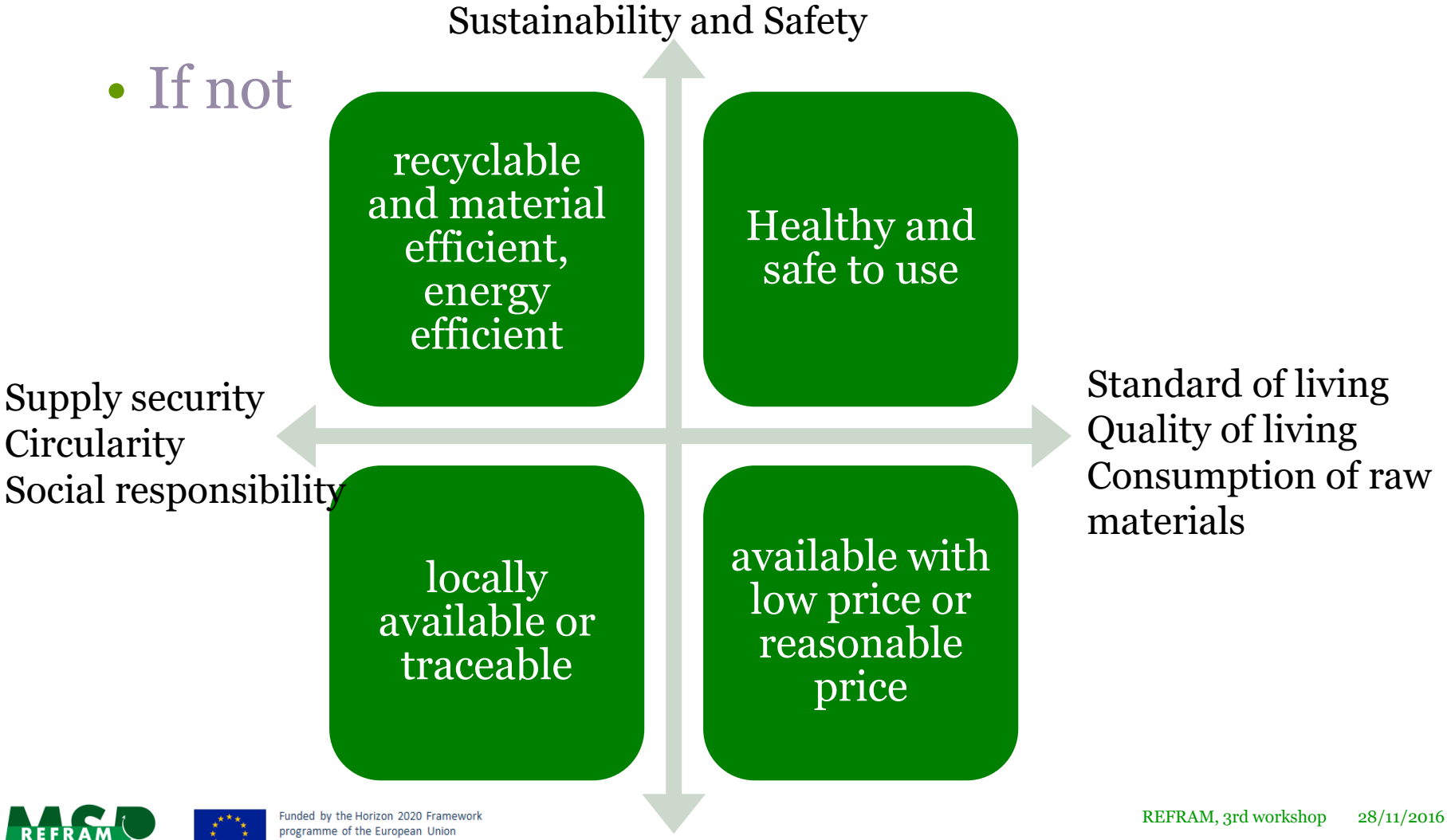
Substitution – WHY?

- If not



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WHAT? to substitute -reasoning and prioritizing

Legislations, politics, funding, mind-set, right players, potential market

READINESS: Ideas TRL,
potential substitutes

PERFORMANCE e.g. **recyclability %, toxic, mechanical properties…**

IMPORTANCE e.g. **application volumes and economic importance**

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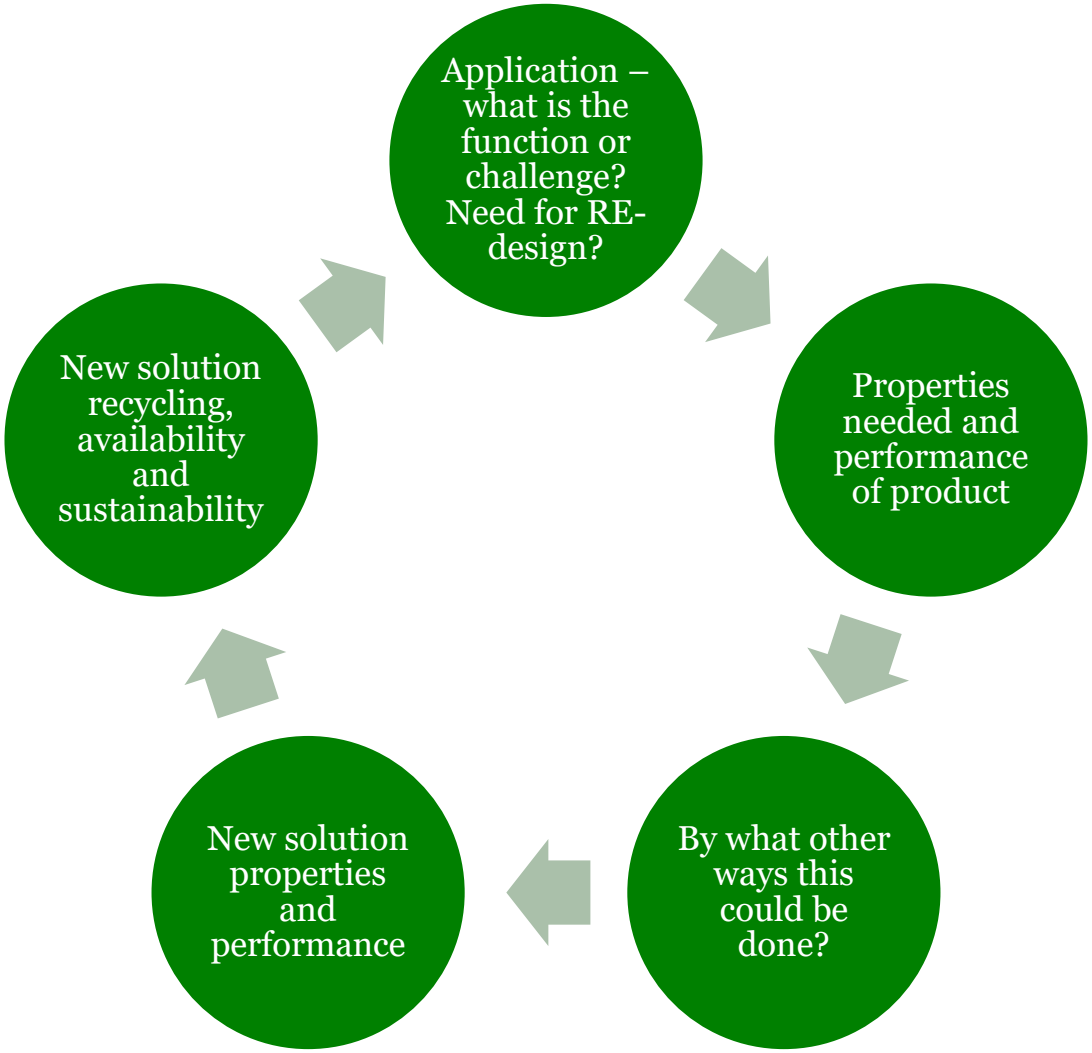
HOW? Design substitutes - By using circular design



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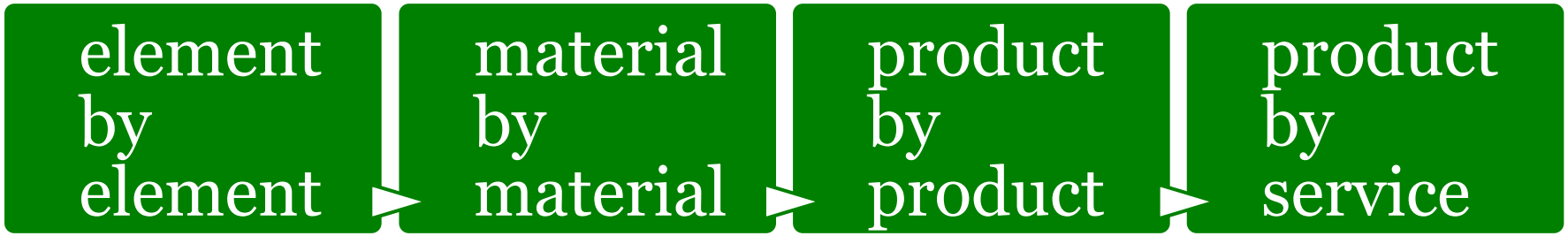
Research and design opportunities

- Substitutes for refractory metals exist but typically research and development is needed.
- Substitutes can be other refractory metals or CRMs (e.g. in steel products)
- Long term research (-10years) investment is needed for technology leap in new material, product or solution development to develop substitutes for refractory metals
- Research of recycled, secondary and un-pure substitutes



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SUMMARY of refractory metal substitution

Refractory metal	Main industrial application	% of usage of current metal	Technical / economical viability for substitution
W	Cemented carbides - WC/Co	72 % of usage in Europe	Mo, Ti
Nb	Steels - HSLA	90 % of usage to steels*	Ti+Mo, V+Mo
Mo	Steels	71%** usate to steels	B, Cr, Nb and V in alloy steels; W in tool steels
Ta	Capacitors	40% capacitors, 21% superralloys	Ceramic, Al, Nb-oxide (capacitors); V, Mo, Hf, Mo, Nb, Re, W
Re	Superalloys	83-90% superalloys***	CMCs

*__USGS Mineral Resources Program.__ *Niobium and Tantalum—Indispensable Twins.* s.l. : U.S. Geological Survey, 2014

**http://www.imoa.info/molybdenum/molybdenum-global-production-use.php International Molybdenum Association.

***Adi Naor, Noam Eliaz, Eliezer Gileadi, Properties and applications of rhenium and its alloys, The AMMTIAC Quarterly, Volume 5, Number 1.

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MOLYBDENUM

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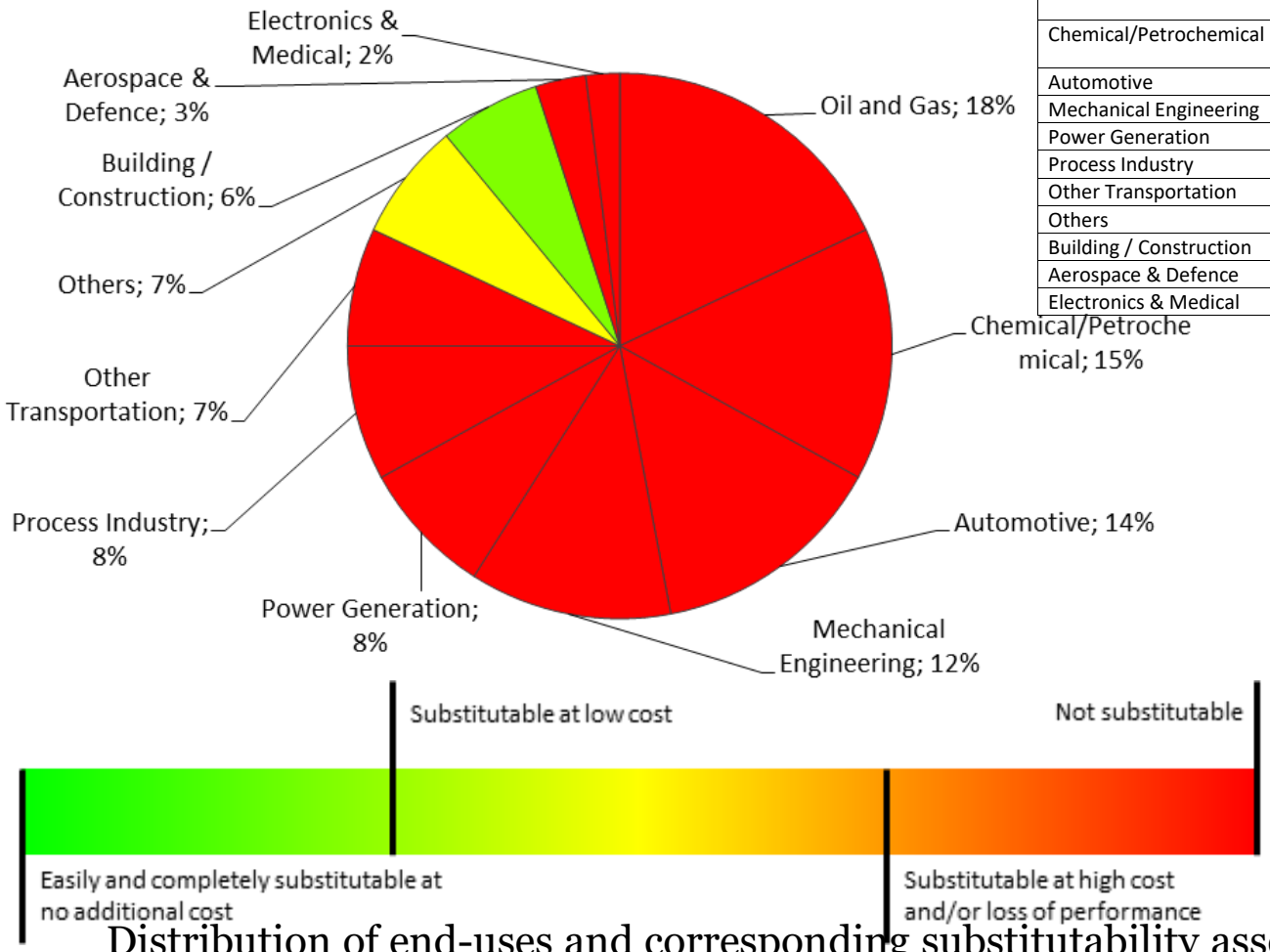
MOLYBDENUM

Substitutability

Application	Share	Megasector	Substitutability index
Oil and Gas	18%	Oil	1.0
Chemical/Petrochemical	15%	Chemicals	1.0
Automotive	14%	Transport-Road	1.0
Mechanical Engineering	12%	MechEquip	1.0
Power Generation	8%	Electrical	1.0
Process Industry	8%	MechEquip	1.0
Other Transportation	7%	Transport-Other	1.0
Others	7%	Other	0.5
Building / Construction	6%	Construction	0.3
Aerospace & Defence	3%	Transport-Other	1.0
Electronics & Medical	2%	Electronics	1.0



Distribution of end-uses and corresponding substitutability assessment for molybdenum

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MOLYBDENUM Reduction potential

- There is little substitution for Mo in major application as an alloying element in steels and cast irons
- Reason - the availability and versatility of Mo lead to development of new materials that benefit from the alloying properties of Mo
- Potential substitutes for Mo include B, Cr, Nb and V in alloy steels; W in tool steels; graphite, Ta, and W for refractory materials in high-temperature electric furnaces.
- For material strengthening and refractory properties - Nb
  - Lower and stable prices
  - Nb is the lightest of the refractory metals
  - Nb is unique in that it can be worked through annealing to achieve a wide range of strength and elasticity

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Source: [4, 10]

MOLYBDENUM Increase potential

- **Alternative material**

application	Current	alternative	advantages	drawbacks
Wear-resistant hard materials	Cemented carbides	ternary borides, e.g. Mo_2FeB_2 or Mo_2NiB_2 particles in metal matrix	equivalent hardness and flexural strength, superior fracture toughness and thermal expansion coefficient	Competitive improvement of cemented carbides – boronizing and other surface protection techniques
Steel protection		Cermet coatings (laser-cladded Mo_2NiB_2)	CET of Mo_2NiB_2 close to steel	Selective removal of Mo_2NiB_2 particles with time
high strength and wear resistance, up to high temperatures,	CMCs	CMCs ductile particle reinforced, e.g. Mo-AlN, Mo-Al_2O_3	Simple production – mix metallic and ceramic powders and sinter; low residual stress	bending strength and fracture toughness depend on Mo content

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MOLYBDENUM Increase potential

- **Alternative system**

application	Current	alternative	advantages	drawbacks
Glass industry corrosion resistant protective coatings	Mo metal sheets	Mo-Re plasma-sprayed coatings	Flexibility of plasma spraying – easy to deposit coating on complex shapes, rhenium improves mechanical properties	Contains expensive rhenium
HT applications, e.g. turbine blades	Ni-base superalloys like CMSX-4	$MoSi_2/Mo_5Si_3$ binary composite	High melting temperature (well above 1000 °C), higher yield stress	

- **Substitution of other elements**

→ in stainless steel molybdenum may up to some point replace chromium – it is a matter of careful assessment of conditions of use, price, estimated lifetime

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MOLYBDENUM Conclusions

- **Molybdenum** production is 50% primary and 50% a by-product of copper mining
- Partially depends on mining economy of major metal (i.e. copper)
- Moderate economic importance
- Non-critical metal
- high level of supply risk strongly connected with low political stability of top producer and top reserve holder
- For most applications (87%) – high substitutability index
- Increase of Mo use at the moment is likely to compromise the performance in most applications, research is ongoing to improve properties

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RHENIUM

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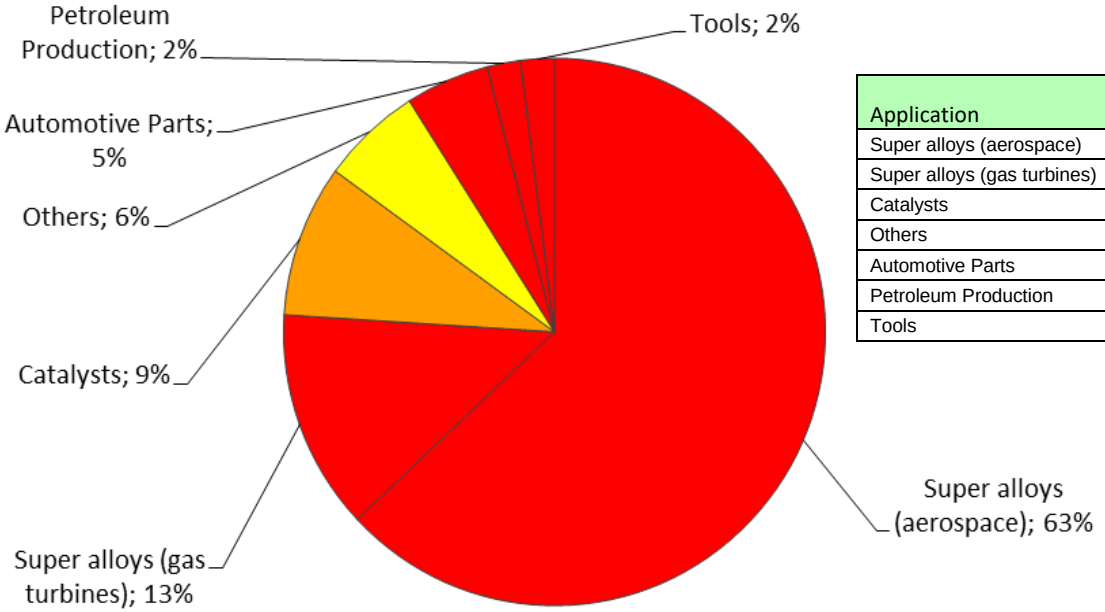
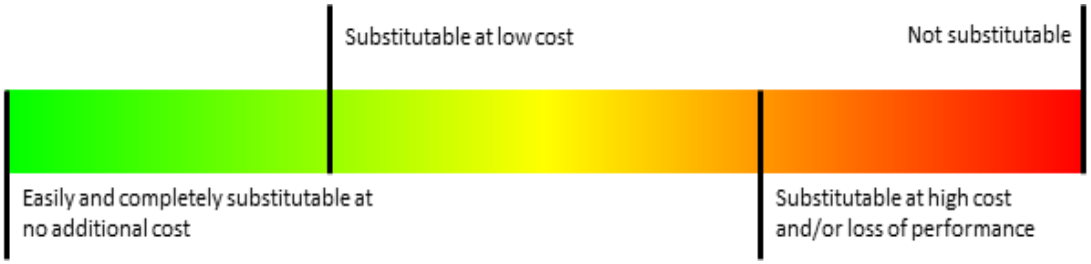
RHENIUM

Substitutability



Application	Share	Megasector	Substitutability index
Super alloys (aerospace)	63%	Transport-Other	1.0
Super alloys (gas turbines)	13%	MechEquip	1.0
Catalysts	9%	Chemicals	0.7
Others	6%	Other	0.5
Automotive Parts	5%	Transport-Road	1.0
Petroleum Production	2%	Refining	1.0
Tools	2%	MechEquip	1.0



Distribution of end-uses and corresponding substitutability assessment for rhenium

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RHENIUM Role in superalloys

- Why Re? → increase HT creep strength of Ni- or Co-based superalloys single-crystal gas turbine engine blades and other components
- Major consumers → aviation
- Accounts for 83% of Re consumption

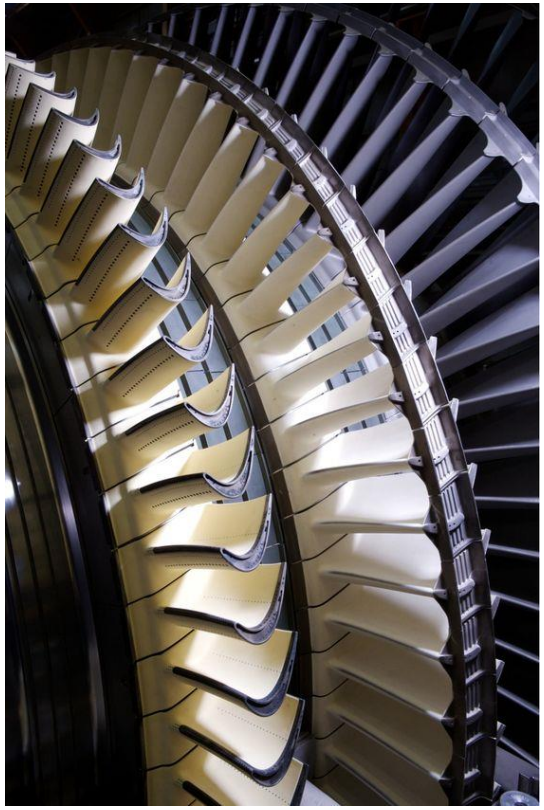

Image 3: Jet engine turbine [https://www.theengineer.co.uk/issues/22-february-2010/turbine-of-the-times/]

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RHENIUM Substitution potential

application	↓↑	Current	alternative	advantages	drawbacks
nickel-based superalloys	↓	CMSX®-2/3	CMSX®-7 CMSX®-8	Improved creep-rupture properties , No or reduced rhenium content	Higher tungsten and tantalum content
Aero engines (high pressure turbines)	↓	superalloys	CMCs	Low density, high fracture toughness, very high resistance to thermal shock	New methods for joining new materials required, due to different heat dissipation in CMCs
HT reactors, fuel cladding	↑	Mg alloys, Zr alloys, austenitic SS	Mo-, W- and other Re alloys	high strength at HT, good mechanical properties at RT	Behaviour under irradiation needed to study
Superhard materials	↑	Diamond-based, borides, nitrides	ReB_2	High hardness	Highly anisotropic properties

↓ - reduction, ↑ - increase

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RHENIUM Conclusion

- ‘Gas turbines are some of the most technically accomplished and complex machines ever created, but despite considerable effort by engine manufacturers there is currently very little substitution potential for rhenium-containing superalloys in the hottest parts of the most powerful gas turbines.’ Roskill Market Outlook Jul 02, 2015

  http://www.prnewswire.com/news-releases/roskill-publishes-its-new-market-outlook-for-rhenium-forecasts-that-rising-demand-for-rhenium-in-aero-engines-will-likely-be-satisfied-by-increasing-supply-of-secondary-rhenium-contained-in-engine-revert-produced-from-end-of-l-511395391.html

- If any – reduction potential is most possible in superalloys – the largest companies already invest in development of materials with reduced Re content - CMSX®-7, CMSX®-8 , CMCs.

- Substitutes for rhenium in Pt-Re catalysts are being evaluated continually. – Ir and Sn have commercial success in one such application. Other evaluated metals Ga, Ge, In, Se, Si, W, V.

- Possible substitutes for Re – Co and W for coatings on copper X-ray targets, Rh and Rh-Ir for high-temperature thermocouples, W and Pt-Ru for coatings on electrical contacts, and W and Ta for electron emitters. Source Granta CES Selector 2015

- Pontential of increase is seen in new applications – superhard materials, fuel cladding materials, but the research is still ongoing

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NIOBIUM

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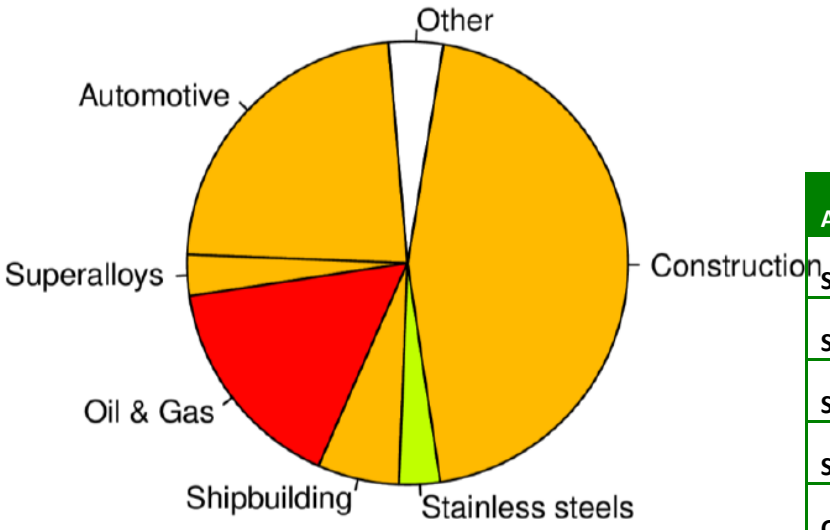
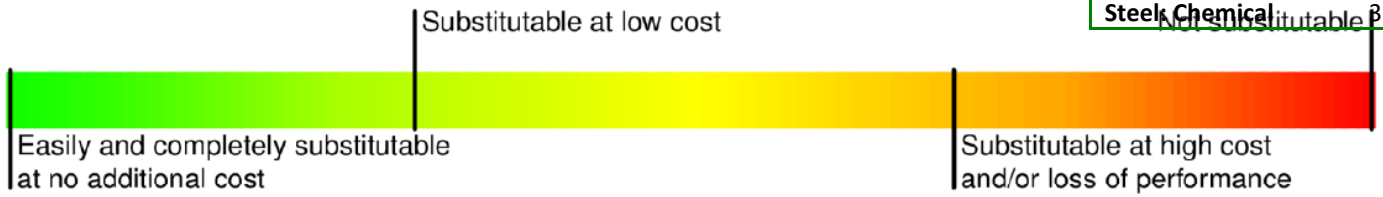
NIOBIUM

Substitutability



Application	Share	Megasector	Value (G
Steel: Structural	31	Construction	104.4
Steel: Automotive	28	Transport – Road	147.4
Steel: Pipeline	24	Oil	50.0
Superalloys	8	Metals	164.6
Others	6	Other	63.3
Steel: Chemical	3	Mechanical Eqpt.	182.4



Distribution of end-uses and corresponding substitutability assessment for niobium

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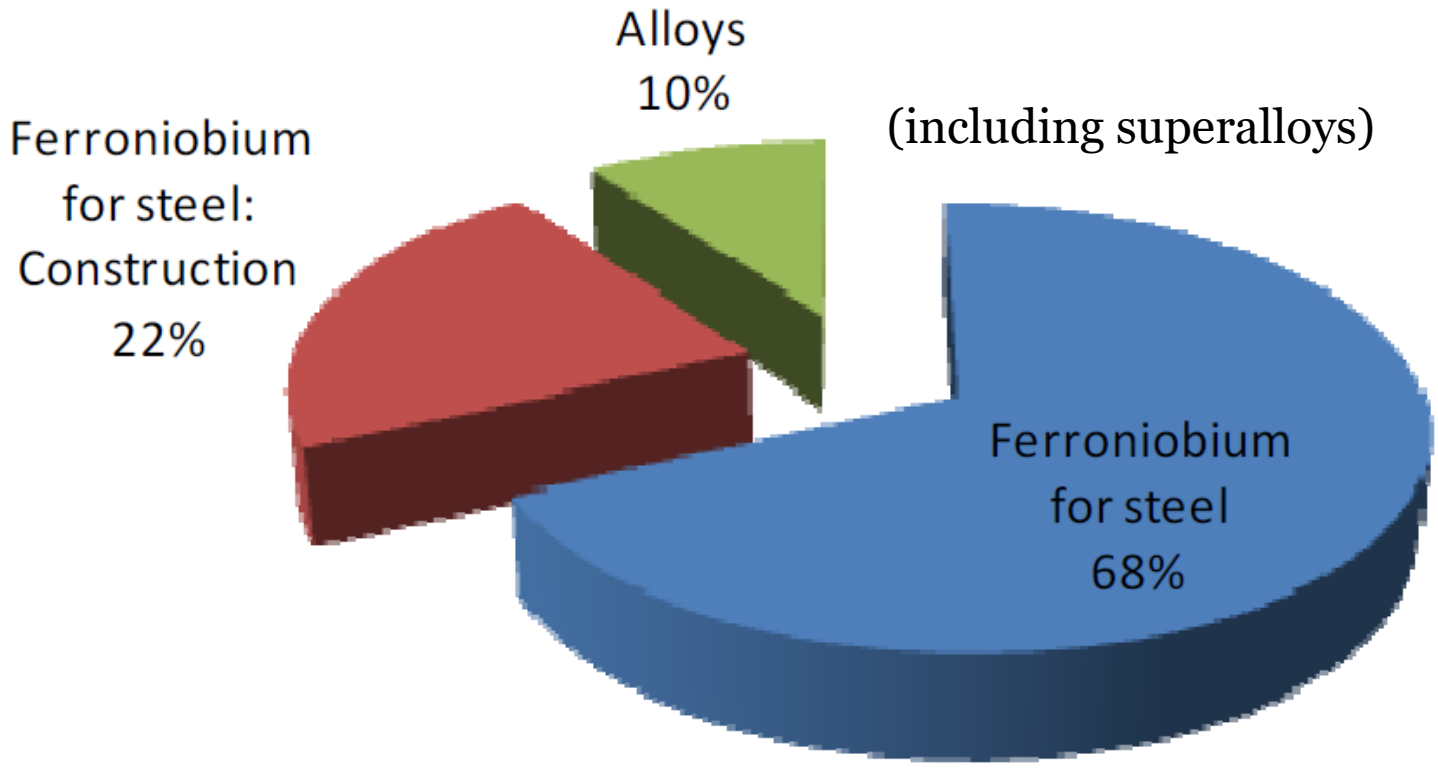
NIOBIUM Consumption share



HSLA steel for automotive applications & pipelines

Image 2 : Niobium consumption & Products [4]

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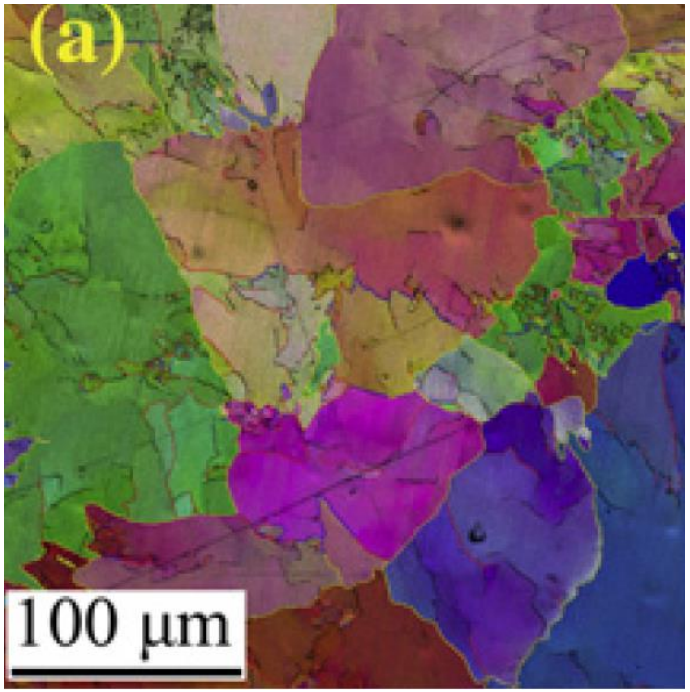
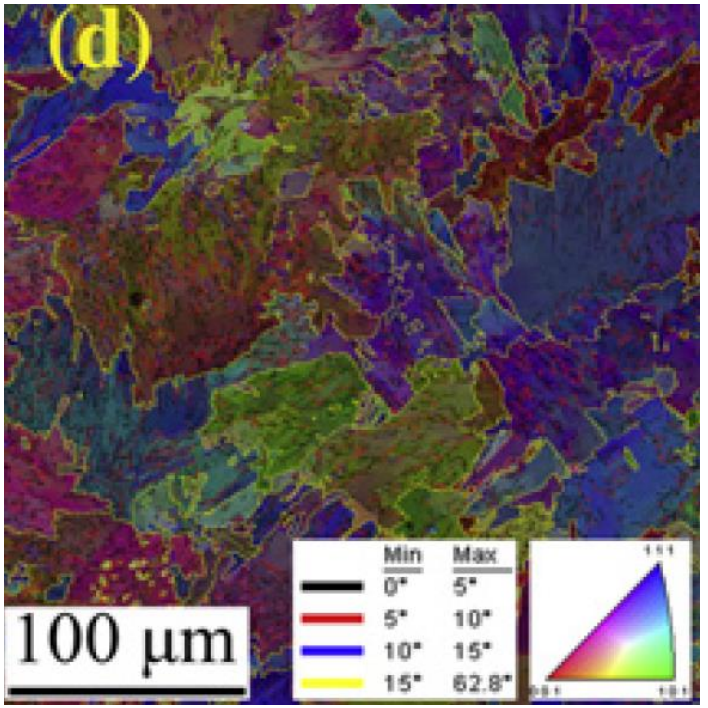
NIOBIUM Role in steels

Decreasing grain size = Increase strength & toughness

Without Nb



With Nb



E. Gözde Dere, Hemant Sharma, Roumen H. Petrov, Jilt Sietsma, and S. Erik Offerman, Effect of niobium and grain boundary density on the fire resistance of Fe–C–Mn steel, Scripta Materialia 68 (2013) 651–654

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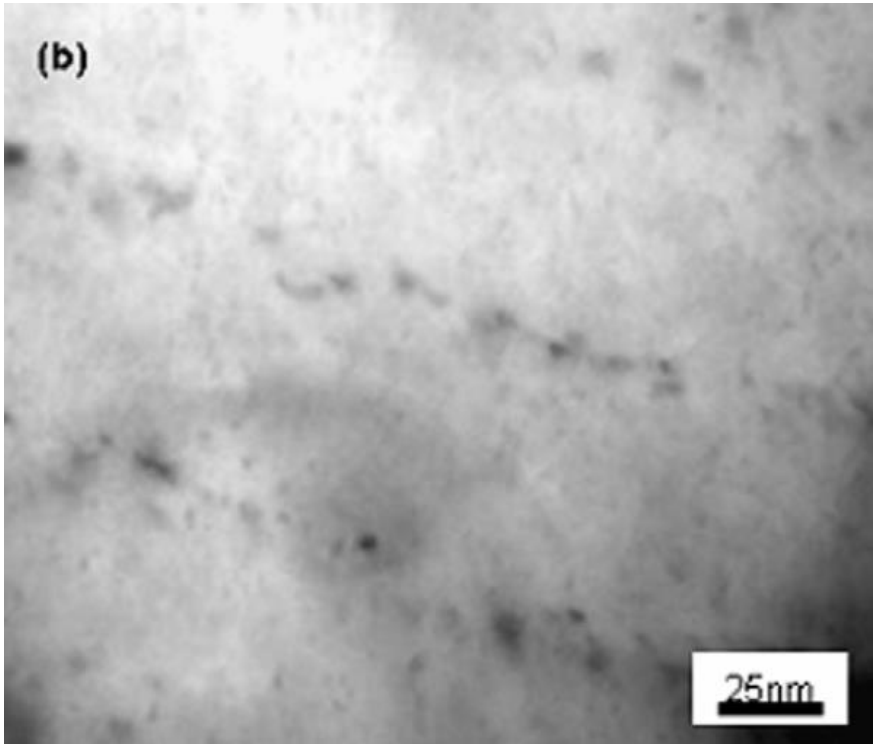
NIOBIUM Role in steels

Precipitation hardening = Increase strength

Nb reacts in steel with Carbon to form nanometer-sized NbC-precipitates



R. Okamoto, A. Borgenstam, J. Ågren, Interphase precipitation in niobium-microalloyed steels, Acta Materialia 58 (2010) 4783–4790

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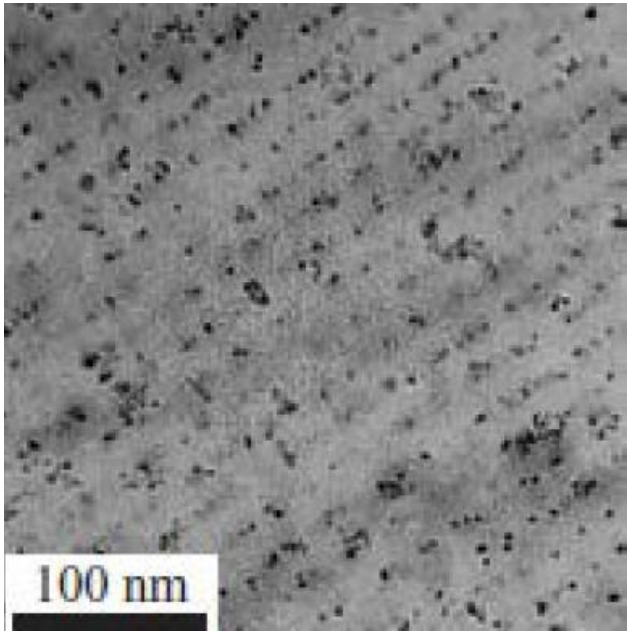
NIOBIUM Potential (partial) substitutes in steel

Titanium & Molybdenum combination: Ti reacts in steel with Carbon to form nanometer-sized TiC-precipitates & Mo keeps precipitates small = increase in strength



Vanadium & Molybdenum combination: V reacts in steel with Carbon to form nanometer-sized TiC-precipitates & Mo keeps precipitates small = increase in strength

Seto, K., Funakawa, Y., and Kaneko, S., "*Hot rolled high strength steels for suspension and chassis parts "NanoHiten" and "BHT" steel*", JFE Technical Report, No. 10, December 2007.

REFRAM

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NIOBIUM Conclusion

- EU consumption of niobium → **24%** of global niobium consumption.
- **No** primary niobium production in Europe → scrap is the only available intra-European raw material source
- Ores and concentrates, oxides and niobium metal → Imported

✓ Need to recycle and find potential substitutes to satisfy increased demand

✓ Potential (partial) substitutes for Nb in steel: 'Titanium & Molybdenum' and 'Vanadium & Molybdenum' combinations

MSP REFRAM

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TANTALUM

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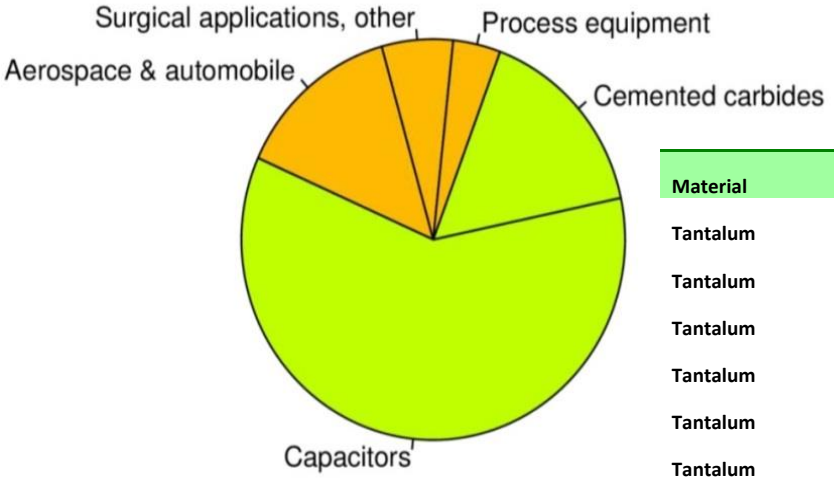
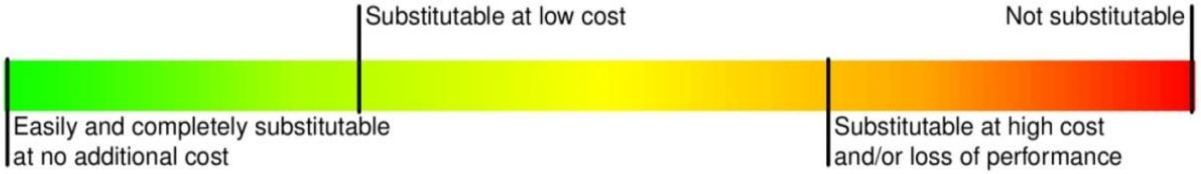
TANTALUM

Substitutability



Material	Application	Share	Megasector	Substitutability
Tantalum	Capacitors	40%	Electronics	**0.3**
Tantalum	Superalloys	21%	Metals	**0.7**
Tantalum	Sputtering targets	12%	Electronics	**1.0**
Tantalum	Mill products	11%	MechEquip	**0.7**
Tantalum	Carbides	10%	MechEquip	**0.3**
Tantalum	Chemicals	6%	Chemicals	**1.0**



Distribution of end-uses and corresponding substitutability assessment for tamtalum

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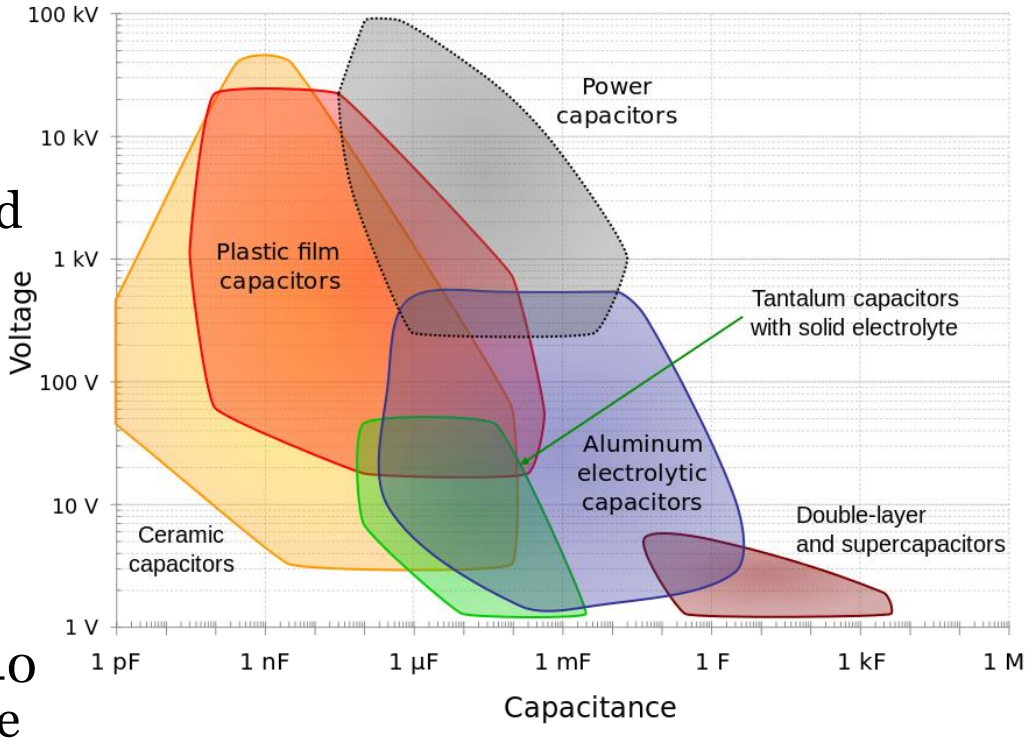
TANTALUM in electronic devices

- High reliability characteristics and low failure rates, operation over a wide temperature range from -55 to +200°C, can withstand severe vibrational forces, small size per microfarad rating/electrical storage capability
- Major consumers in electronics industry, most of it is used in powder or wire form in manufacture of **capacitors**
- Accounts for about 60% of the yearly Ta consumption, about 40 % (of total in 2011) manufacture of capacitors



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TANTALUM in superalloys

- Alloy compositions containing 3-11% tantalum (typically nickel based) offer resistance to corrosion by hot gases, allow higher operating temperatures and thus efficiency and fuel economy
- Major consumers in **aerospace** (75% of super-alloy demand, including jet engine and rocket engine nozzles) and **defense** applications (e.g. missile parts), also suitable for turbine-type equipment, such as gasturbines
- About 14% to 21% of yearly consumption of Ta.

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TANTALUM in cemented carbides

- Increased high temperature deformation, control of grain growth, increases thermal shock resistance and reduces high temperature oxidation of the tools,
- Typically contain about 3% tantalum
- Major consumers: high-speed **cutting and boring tools**, and other tools for environments with high levels of stress and temperatures (e.g. teeth for excavator buckets, mining drills, high-performance bearings and cutting blades).
- Tantalum carbide (TaC) accounts for about 12-14% of yearly consumption of Ta.

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TANTALUM Potential substitutes

The following materials can be substituted for tantalum, but usually with less effectiveness:

- **aluminum and ceramics** in electronic capacitors;
- **niobium** in superalloys and carbides;
- **niobium, glass, platinum, titanium, and zirconium** in corrosion-resistant equipment;
- **niobium, hafnium, iridium, molybdenum, rhenium, and tungsten** in high-temperature applications.

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Sources: [4, 9, 10, 11]

TANTALUM Conclusions

- At present there are **no** primary production of tantalum in Europe
- Drivers for substitution (criticality, price, recycling rate) show there are clear need for substitution or increasing recycling rate
- Tantalum can be substituted by other materials but most substitutes have higher costs or adverse properties.
- For Tantalum main application, capacitors, possible substitutes exits (by aluminium and ceramics) and are likely to answer most common needs.
  - Ta capacitors are still used in applications which require high performance
- In other application areas possible substitution of tantalum by niobium (**CRM**) is possible – rationale?
  - In cemented carbides also titanium carbides (TiC) and nitride (TiN) are possible
  - In corrosion-resistant equipment: glass, platinum, titanium, and zirconium
  - In high-temperature applications: hafnium, iridium, molybdenum, rhenium, and tungsten

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TUNGSTEN

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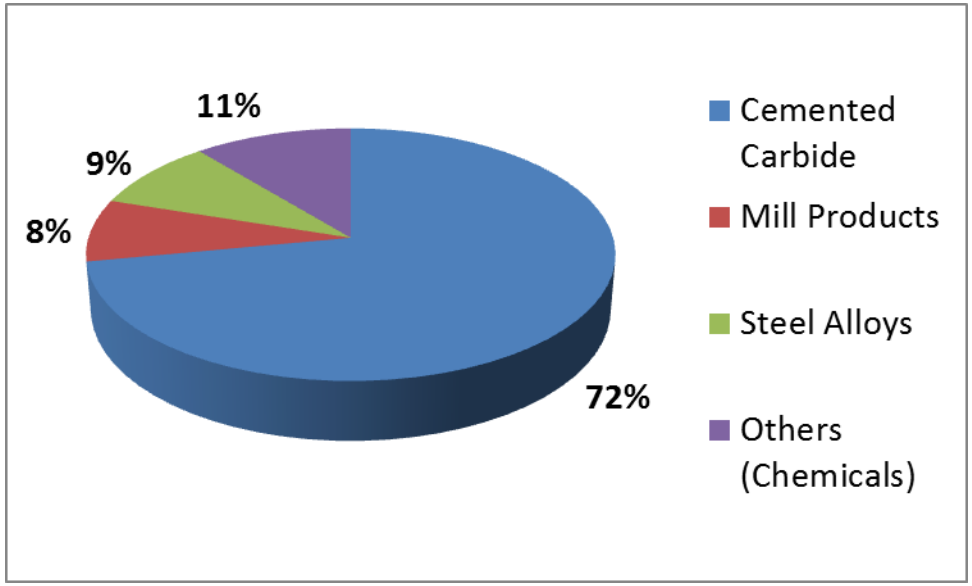
Applications

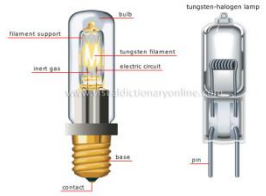
Application of Tugnsten in EUROPE



Hardmetals-cutting materials manufacturing

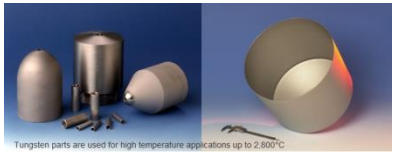
Mining and Construction

Lamp industry



High-speed steels

High temperature applications



X-ray tubes

Turbine blades

Catalysts

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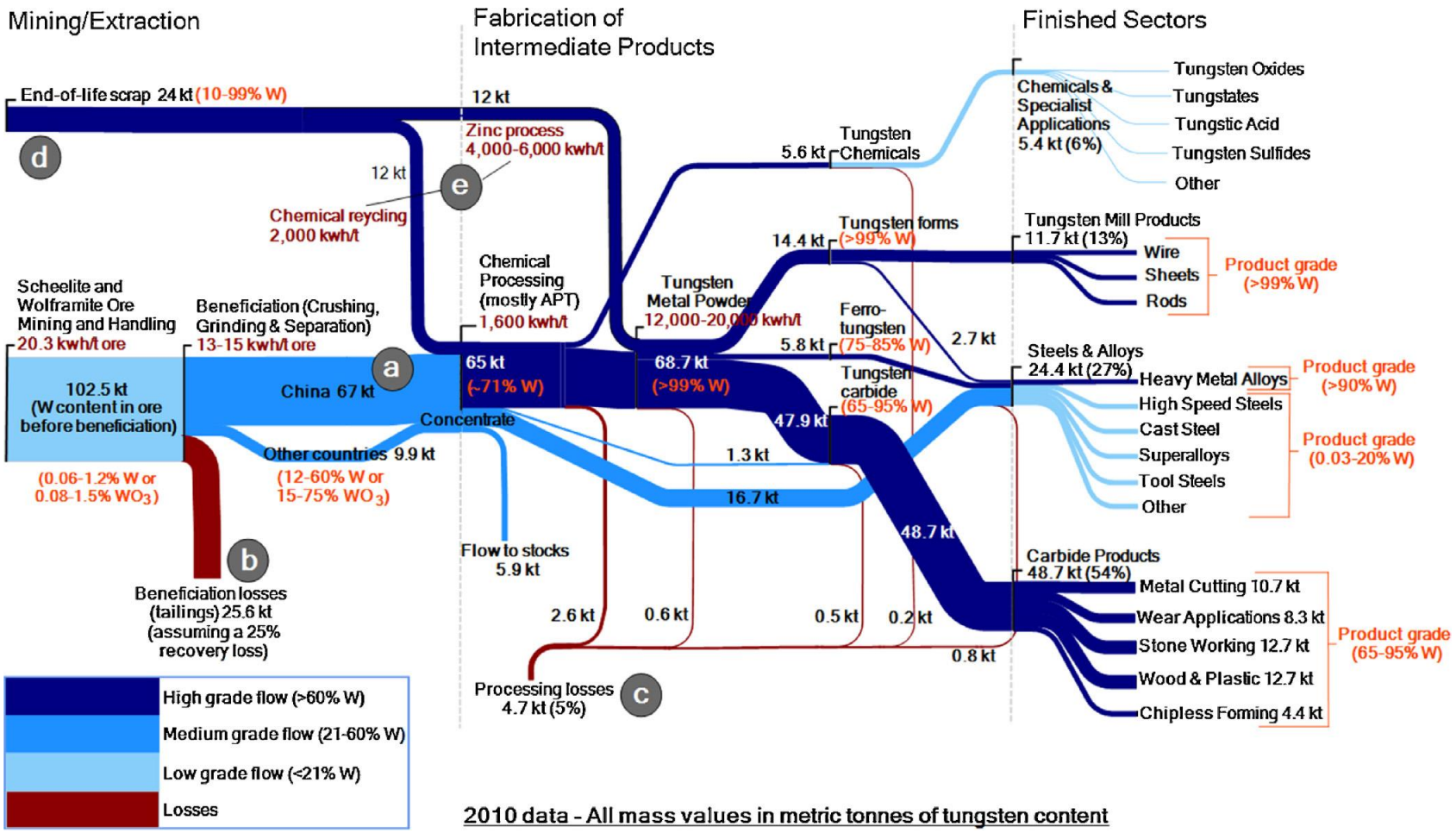
Global mass flows of tungsten in 2010



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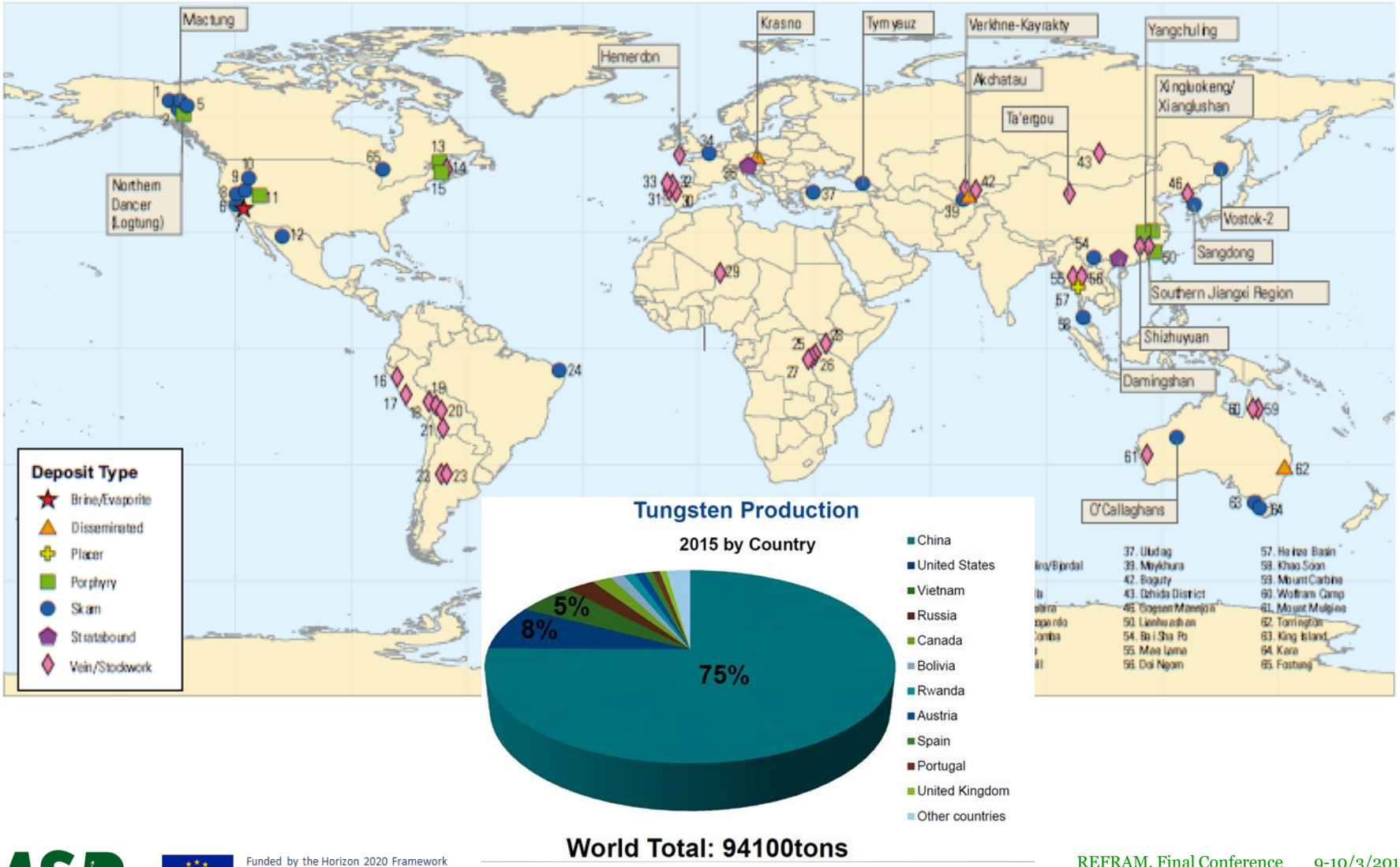
Global distribution



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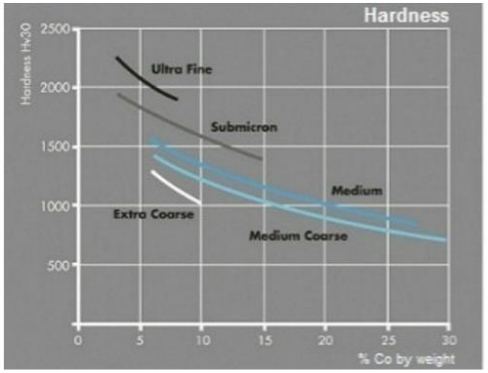
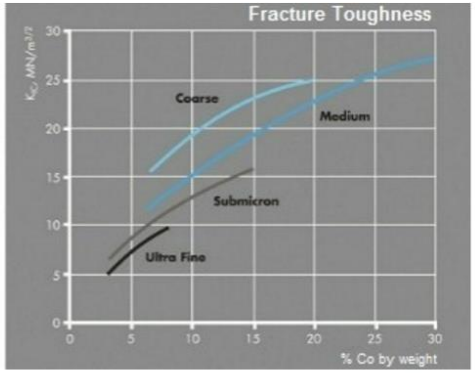
TUNGSTEN Role in Cemented carbides

- **Cemented carbides (hardmetals)**→the most important usage of tungsten today. The main constituent is tungsten monocarbide (WC).
- Cemented carbides are materials made by "cementing" very hard **tungsten monocarbide (WC)** grains in a binder matrix of a tough cobalt or nickel alloy by liquid phase sintering. The high solubility of WC in the solid and liquid cobalt binder at high temperature provides a very good wetting of WC and results in a excellent densification during liquid phase sintering and in a pore-free structure.
- WC play a crucial role as they combine high **hardness** and **strength** with **good toughness** within a wide property range, and thus constitute the most versatile hard materials group for engineering and tooling applications.





Hardness increases with decreasing grain size and binder content.
Hardness increases fracture toughness decreases and vice versa.
In truly abrasive applications, hardness is a good measure of wear resistance.

- Cemented carbides are their high modulus of elasticity (about three times that of steel), their extremely high compressive strength (up to 8,000 MPa) making them ideal tool material for high pressure diamond synthesis, and an excellent thermal conductivity.

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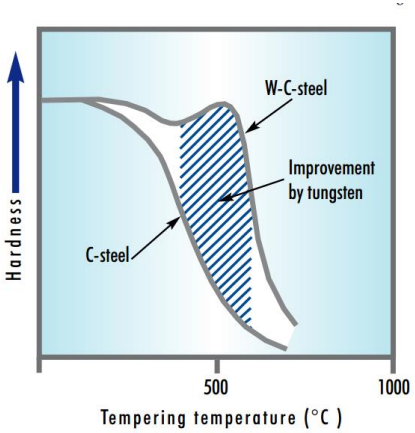
Role W in Steel Alloys

- Tungsten significantly improves the **hot hardness and hot strength** of steel and leads to increased yield strength and tensile strength without adversely affecting ductility and fracture toughness.
- **Alloying element in steels**: High speed steels, hot work tool steels, cold work tool steels, plastic mould tool steels, heat and creep resistant steels, corrosion resistant stainless steels and valve steels.
- Tungsten has the **highest melting point** of all metals and is alloyed with other metals to strengthen them. Tungsten and its alloys are used in many high-temperature applications, such as arc-welding electrodes and heating elements in high-temperature furnaces.
- Tungsten in steel increase the amount of undissolved and excess carbide in the hardened steel and the eutectoid points is shifted towards lower carbon concentration. The consequence is the precipitation of fine or very fine grained carbides evenly distributed in the steel matrix.
- **Good wear resistance** is the most important property of tool steels, and tungsten, next to vanadium, is the most effective carbide forming element for increasing wear resistance.

Early history of high speed steels

year	C	Cr	W	Mo	V	Si/Mn	Co	remarks
1868	1.5-2.4	0.5	**5.0-8.0**	-	-	~1.0/2.0	-	Mushet; first "self-hardening" steel
1886	2.0	12.0	×			×		Brustlein; J.Iron Steel Inst. 1886
1898	1.8	3.8	**8.0**	-	-		-	Taylor-Wight ("overheated")
1905	0.25-2.0	1-7.0	**4-30.0**	2-15.0 alt. to W	0.1-1.0			Mathews US 779,171
1906	0.68	5.95	**17.81**	-	0.32			Taylor-Wight
1910	0.8	4.0	**18.0**	-	1.0		-	McKenna
1912	0.8	4.0	**18.0**	-	1.0		5.0	addition of Co
1923	0.8	4.0	**18.0**	-	1.0		12.0	
1939	1.4	4.0	**18.0**	-	2.0-5.0		-	high C, high V
1940	×	×	-	9.0	×		×	W replaced fully or partially by Mo
	×	×	**6.0**	5.0	2.0		-	



The effect of tungsten alloying on hardness during tempering

Fig.1: Effect of tungsten on hardness after tempering (schematic); [1] hardening is achieved by austenitization and subsequent quenching and multiple tempering at ~550°C to obtain a fine, mainly martensitic steel matrix with precipitated very fine alloy carbides (secondary hardening)

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Role W in Super-Alloys

- **Super-alloys→** High temperature strength, high thermal fatigue resistance, food oxidation resistance, excellent hot corrosion resistance, air melting capability.
- W alloyed nickel- and cobalt-based super – alloys are used in aircraft engines, marine vehicle, turbine blades and vanes, exhaust gas assemblies and as construction material for furnace parts

Role W in Mill products

- **Mill products→**Extremely high melting temperature (3414 ºC), low vapor pressure, high stiffness and excellent creep resistance at elevated temperature
- W used in the form of wires, coils and coiled coils incandescent lamps and as electrode in low-and high pressure discharge lamps

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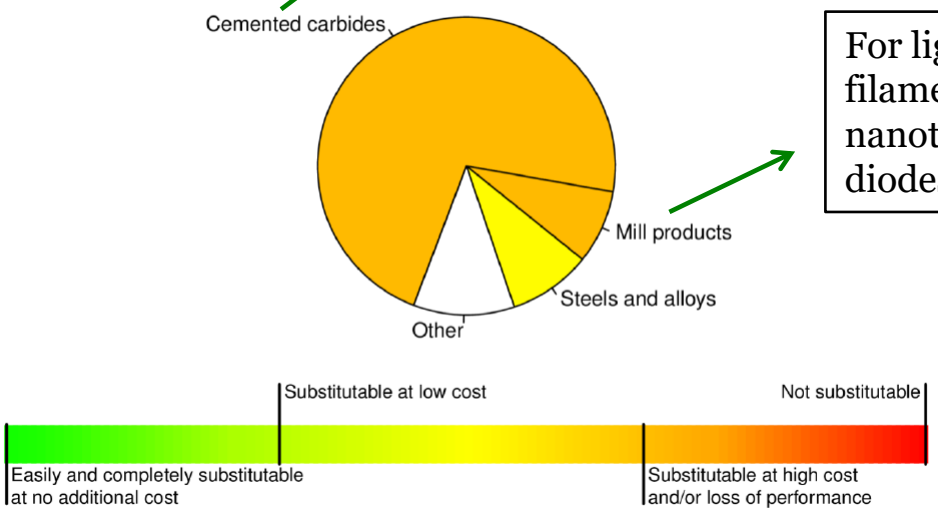
Substitutability

Substitutes for most application results in a loss of performance or in an increase of cost.

In cemented carbides and other applications, W and its compounds can be replaced by Mo, Ti, ceramics depleted uranium or hardened steel.



For lightning equipment tungsten filaments can be substituted by carbon nanotube filaments, light-emitting diodes or other light sources.

Distribution of end-uses and corresponding substitutability assessment for tungsten

Figure 214: Substitutability of tungsten scoring used in the analysis

Use	Substitutability score
Tungsten alloys	0.7
Superalloys	1.0
Fabricated products	1.0
Alloy steels (mainly tool steel, >80%)	0.7
Cemented carbides	0.7

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TUNGSTEN

Summary

- The consumption of W continues to increase as the amount of carbides tool production increases with the expansion of markets in developing countries.
- For Tungsten main application, **WC-based cemented carbides, since to be difficult to substitute: increase of cost and lower performance**
  - Titanium carbides (TiC) and nitride (TiN) **are potential substitute but the technology is not competitive at the moment**.
- In other application areas possible substitution of Tungsten is possible – rationale?
  - Super-alloys substituted by Ceramic matrix composites (CMCs) made from a silicon carbide/nitride matrix for gas turbine engines. In some applications possible. Nanostructured n-alloys possible (but TRL is still 3-4) – 10 y scenario.
  - Difficult to replace in some mill products. Already substitution path in LEDs
  - Steel products can be replace by other CRM (Nb) or refractory metals (Mo). New perspectives via GB engineering and new alloys.

REFRAM

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Source: [4, 10]

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DISCUSSION

THANK YOU!

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Source: [4, 10]