Computational Multi-Models Enabled Design of Safe & Sustainable Multi-Component High-Entropy Coatings

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Abstract. The project M2DESCO is directed at addressing new challenges for developing next-generation high-entropy-alloy-based multi-component green (free of toxic substances) and sustainable (rare earth free & minimum critical metal elements) coatings with predictable functionalities, performance and life - aiming at increasing wear resistance by 100%, corrosion/oxidation resistance by 50~60 %, and effectively reducing the criticality of coating materials by at least 70%. To achieve these goals, the project is to integrate artificial intelligence/machine learning AI/ML underpinned, highly effective and highly efficient Computational Modelling that is guided by a novel Safe and Sustainability by Design Framework and facilitated by high-throughput characterisations and evaluations, to speed up the material-design to coating-product development process (reducing the development cycle-time by 400~500%). At the same time, M2DESCO is to enable optimal alloy/material design and coating process optimisation for high efficiency and high quality, so to reduce the overall product manufacturing cost (due to use of the new tooling developed) by 20%. The advancement of M2DESCO will contribute significantly to combating the loss in EU region caused by corrosion and wear, to the enhancement of the global profile and leadership of the EU material modelling/research

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community, to strengthening of the innovation capability of the EU coating industry/business.

1 Introduction

The ever-increasing demands for high-performance, high-efficiency, reduced energy-consumption and increased service-life of mechanical equipment and tools operated under extreme environments (such as cutting tools for highspeed dry machining and hot forging dies) have been the major drivers for developing advanced coatings to combat the premature or even catastrophic failures of mechanical equipment and tools due to corrosion, wear and oxidation.

The past two decades have witnessed rapid developments of surface coating technologies [1] but it ends up with more and more uses of critical metal elements (such as Nb, Co, V, W, in most coatings for tools, and also in tool materials) and continue use of toxic substances (such as Cr+6 for hard Cr coating) for some demanding applications (such as actuators for landing gears), due to lack of satisfactory green coating replacements. This today situation is illustrated on the top-right of the sustainability-safety risk diagram of Fig.1.

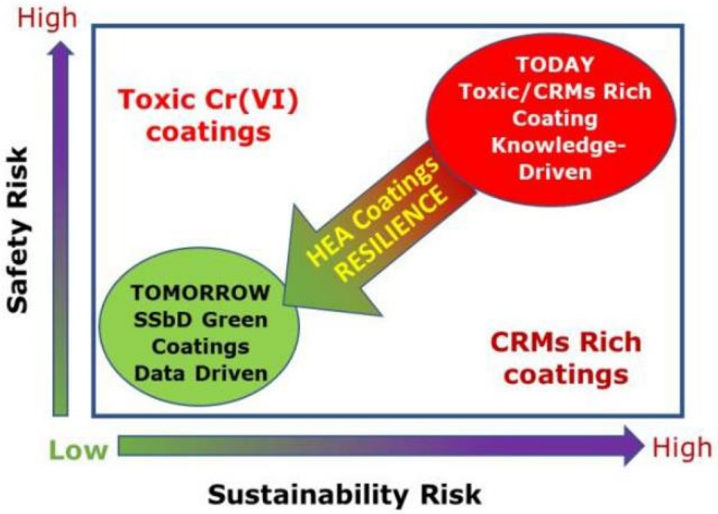

Fig. 1. Scheme of present (TODAY) and envisaged (TOMORROW) situation for the industrial design of industrial coatings, as positioned in a sustainable-safety risk map.

High Entropy Alloy (HEA)-based multicomponent coatings (MCHECs), including HEA coatings and High Entropy Ceramic (HEC) Coatings, are emerging as next generation of coatings with outstanding combinations of mechanical (high hardness and toughness), chemical (high resistance to oxidation, corrosion and sticking/adhesion), physical (high thermal stability & low diffusion) and tribological properties (high wear resistance), which are unattainable by currently available coatings [2-3]. However, most HEAs and MCCs contain a large amount of critical metal elements (CMEs) such as Nb, Co, Ta, W, V etc. with high criticality index values [4], in spite of that there are a high degree of freedom in alloy design with, in total, 108 combinations of 64 potential elements.

Currently, the development of MCHECs is mainly based on mere experimental approaches, which significantly restricts the number of possible exploratory combinations.

This is largely due to lack of efficient and accurate computer-based multi-models (= Multi-scale and Multi-physics + Data-driven + safety and sustainability assessments/predictive models) and support software which are able to predict basic properties as a function of the coating compositions and processing conditions. The current modelling largely relies on individual (rather than integrated multi-models) empirical models, and the models are unable to deal with, successfully, non-equilibrium PVD deposition processes and non-equilibrium phase diagrams of the MCHEC substances, being all with highly disordered structures & complex properties. To date, most computational models deal with bulk HEAs and these are applied separately from the film-growth structures, not giving adequate considerations on the safety, risks and lifecycle sustainability related issues, not mentioning the tools empowered with advanced AI/ML algorithms that could substantially reduce the computational time, and hence, the design cycle-time. These limitations have restricted current effort in searching for optimal alloys to a limited number of trials. Therefore, the potential for using MCHEC for creating high-quality and high-performance coating cannot be explored fully up to now. As a result, it is very difficult, if not impossible, to design and develop HEA-based coatings with desirable functionality, low life-cycle environmental impacts and high circularity. Thus, the EU coating material (especially in the case of hard coatings) and manufacturing industry is unable to explore more opportunities to enhance their contributions to the EU green deal and to achieve industrial resilience to the extent which the industry would like it to occur, according to the red-to-green evolution depicted in Fig. 1, for tomorrows coating industry.

2 Research themes and methodologies

Fig. 2 illustrates the development pipeline of the project to evolve from lower technology readiness levels (TRLs) to higher ones towards eventually commercialisation and associated timeline.

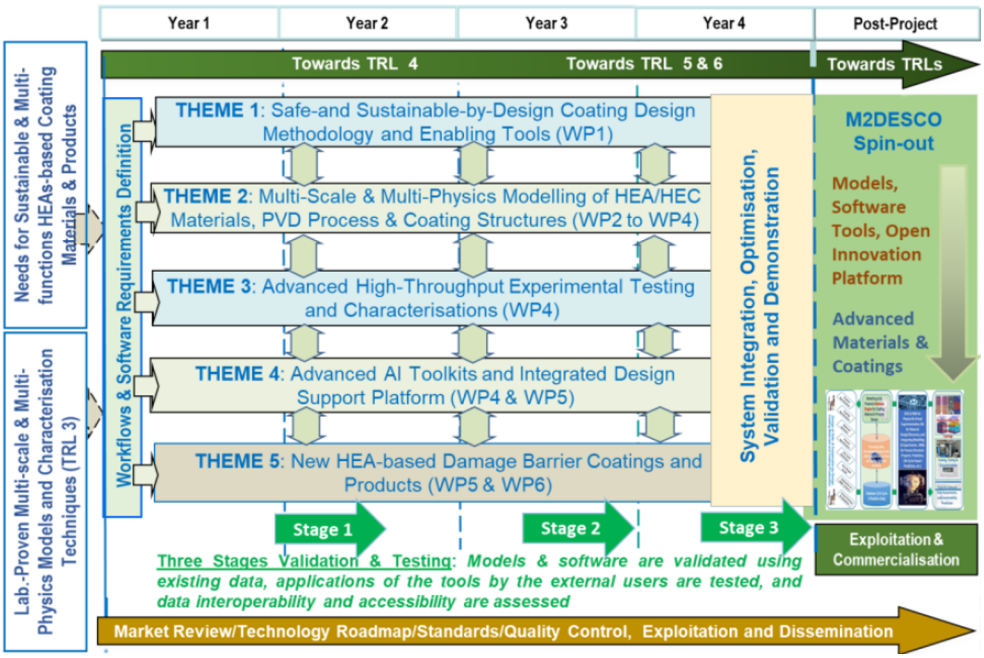

Fig. 2. M2DESCO technical pipeline, including scientific research themes as well as the stages for validations and the envisaged exploitation targets.

M2DESCO gather 5-highly innovative comprehensive research lines (denoted as Themes) as depicted in Fig. 2: i) safe and sustainable by design (SSbD) coating designs and methods, ii) multi-scale multiphysics modelling of HEA/HEC coatings, mainly synthesised by the Physical Vapor Deposition (PVD) techniques; iii) high throughput experimental testing methods, iv) advanced artificial intelligence (IA), and a purpose-built platform to support coating design, gathering all the previous inputs. Finally, a Theme iv) will be carried out to launch and validate new families of damage barrier coatings for industrial products.

3 The multi-scale models and their interconnections

The nucleus of the project activities and its success lay in the development of a comprehensive multi-scale physical model in which efficient predictive-tool, accelerated through AI and ML can act on specific materials size scales and receive-provide the correct inputs-outputs in an efficient way. In order to illustrate this concept, Figure 3 shows a scheme of the multi-scale framework envisaged at M2DESCO.

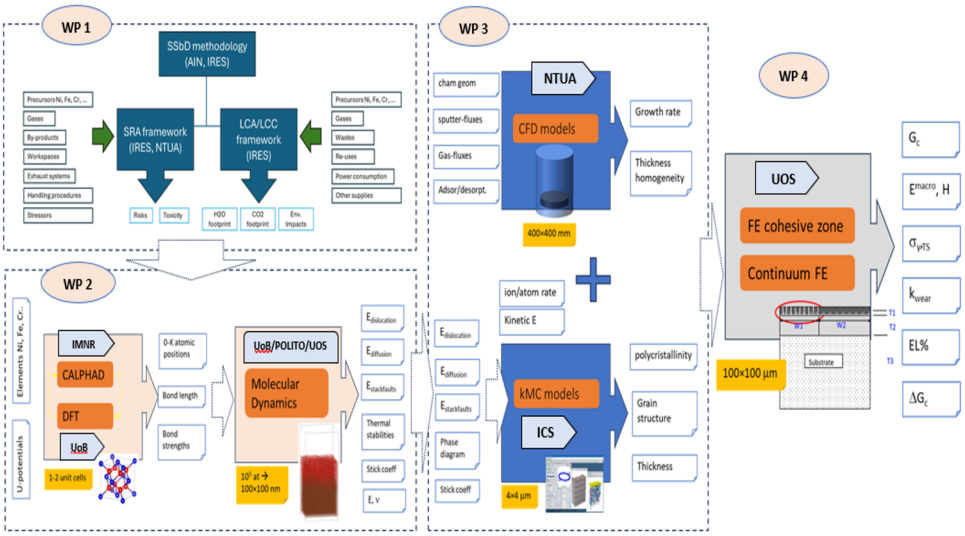

Fig. 3. Scheme of the interconnected multiscale models for HE coatings.

The multiscale model consists of 4 main interconnected blocks. The block WP1 gathers all the necessary methodologies to carry out a coating materials selection on the basis of SSbD and safe-risk assessment (SRAs), including a thorough Life Cycle Assessment. As inputs, the precursors of the coatings, gases used during processing, by-products, handling, power consumptions, wastes and other stressors are provided. As outputs, toxicity-risk-criticality indexes, and CO2/H2O footprints are expected.

The block WP2 covers the physical modelling to estimate basic coating properties, such as crystal structure stabilities, bond strengths, elasticity, and thermal properties; by coupling equilibrium phase diagram calculations, using CALPHAD and Density Functional Theory (DFT) [5]; and thermomechanical properties by Molecular Dynamics (MD) [6]. Advanced MD such as reactive-MD shall also provide valuable model outputs such as friction, diffusivity of atomic species, wetting, fracture toughness and plasticity of crystal structures, all being fed to the next blocks for larger size scale models (3 and 4).

Block WP3 encompasses out-of-equilibrium process simulations using computational fluid dynamics (CFD) to replicate the coating process within an industrial PVD reactor, and

at atomic level (using up to 106 atoms) the so-called kinetic Monte-Carlo simulations of film growth, to reproduce polycrystalline grain growth, and defects. To this aim, novel augmented versions of the commercial software 'Virtual Coater' by ICS (Belgium) shall be developed to account high ionized vapor plasmas as characteristics of arc evaporation and high impulse power magnetron sources (HIPIMS).

Finally, the block WP4 will receive as input parameters all these produced by the models 1 to 3 and produce at larger scales (10-100 microns) thorough models to predict the coating/substrate system behaviour under working conditions, such as cyclic mechanical loading or thermal stresses.

4 M2DESCO industrial cases

M2DESCO will evaluate the impacts of the outcomes on 2 specific manufacturing processes, by intercomparing the results of the multi-scale models with specific coating production and testing. The uses cases encompass i) the manufacturing of metal parts by hot forging and ii) the machining of materials difficult to cut such as titanium and Ni-super alloys, hardened steels, and metal matrix composites for different parts of electric vehicles; turbines, transmission gears, break-paths, etc.

A range of surface coatings have been developed to protect against abrasion, oxidation, corrosion, thermal fatigue and work material soldering (galling) etc.[3]. However, extremely hard coatings are normally characterised by low ductility and hence, low damage-tolerance, mainly due to their complex structure with brittle phases. High temperatures also affect the coating resilience due to oxidation and hard phase decomposition, as induced by atomic diffusion processes. It is in this context where emerges the high interest in HEAs as a new generation of metallic coating materials with a unique combination of attractive mechanical (desirable combination of high hardness and toughness), chemical (high resistance to oxidation, corrosion and sticking/adhesion), physical (high thermal stability & low diffusion) and tribological properties (high wear resistance),

The coatings to be implemented will be developed by the PVD technology using industrial systems equipped with up to 4 magnetron sources which allow to carry out DC-pulsed, dual Bipolar or HIPIMS sputtering modes. PVD is a well-established key enabling technology which provides value to numerous industrial and consumer goods. Figure 4 illustrates some of the products where PVD coatings can be applied, from metal tools (cutting and forming) to applications in decoration, packaging, solar energy conversion, optics, house-holds, H2 production by electrolysis, H2-fuel cells, or even medical inserts.

The production of experimental coatings will be carried out by combining multi-compositional target approaches. In this mode, high throughput (HTP) experimental production can be attained (i.e. different coating formulations can be produced in one single batch). Besides, the HTP experimental production concept shall also be applied to obtain large characterization datasets, for example nano-indentation, chemical profiles, or crystallographic/microstructural properties, which can later be used to feed AI/ML algorithms to qualify the results obtained through the multi-scale models.

5 Conclusions

M2DESCO consortium sets up ambitious targets in developing advanced algorithms, multi models and a series of software tools to support the development of novel Multi-Component High-Entropy Coatings that are safe and sustainable, assisted by the development of advanced safety and sustainability assessment framework, underpinned by a Safe and Sustainable-by Design methodology, in addition to the development of AI/ML toolkits,

collaborative platform, fast material testing and characterisation techniques and protocol. These will be validated through several industrial test cases. The aim is to increase wear resistance of the targeted coatings by 100%, corrosion/oxidation resistance by 50~60 %, and effectively reduce the criticality of coating materials by at least 70%, while these are to be achieved with much shortened development cycle-time.

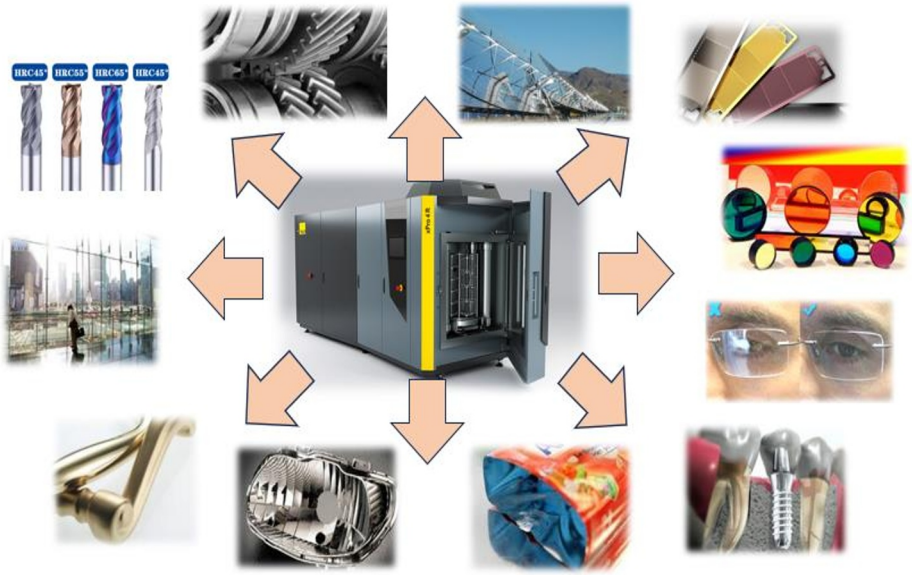

Fig. 4. Value added by PVD technology (central image) to a series of industrial and consumer good products.

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