Characterization of wall response to plasma fluctuations in tokamak

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Plasma boundary conditions in SOL provided by dynamic hydrogen recycling from PFCs governed by complex and multi-faceted material processes

- Hydrogen retention and recycling in PFCs governed by various complex and multifaceted atomic processes in material bulk and on material surface

- Various plasma regimes in Tokamak scrape-off layer with transient features (fluctuations, ELMs, ...): PFCs exposed to a broad range of plasma conditions fluctuating in time

- Example of synergetic effects between plasma and H recycling: Re-healing of giant ELMs determined by wall outgassing, which controls giant ELMs frequency

- Integrated modeling of PMI using macroscopic plasma models and atomic wall models virtually impossible due to the multiscale nature of PMI

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Reduced continuum models of hydrogen retention and recycling (e.g. FACE\(^1\), Xolotl\(^2\), ...) must be developed to describe dynamic wall recycling and provide time-dependent boundary conditions for plasma solvers (SOLPS, BOUT++, ...)

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\(^1\) R. Smirnov Fusion Sc. Tech. 2017 \(^2\) S. Blondel FTS 2017 \(^3\) A. Pigarov JNM 2014
Characterization of wall response using reaction-diffusion models in conjunction with first-principle atomic modeling

- Splitting between plasma models and wall models possible when considering linear wall & plasma responses\(^1\)

\[ \Gamma_{in}, Q_{in} \]

\[ \text{core plasma} \]

\[ \text{SOL plasma} \]

\[ \text{plasma} \]

\[ \Gamma_{in}, Q_{in} \]

\[ \text{Wall response} \]

\[ W(\Gamma_{in}, Q_{in}, c_k, T) \]

\[ \Gamma_{out}, Q_{out} \]

\[ \text{long-term retention} \]

\[ c_k, T \]

- Fully coupling plasma and wall models however requires to develop an advanced numerical framework → **we focus solely in this work on the analysis of the wall response**

- Atomic and macroscopic modeling of H transport, reaction and desorption processes must be combined to describe hydrogen recycling from PFCs (wall response)

\[ \frac{dc_H}{dt} = \text{transport} + \text{trapping} + \text{detrapping} + \text{source} \]

\[ \frac{dc_s}{dt} = \text{desorption} + \text{bulk ↔ surface} \]

**First-principle atomic modeling of material**

\(^1\) S. Krasheninnikov PoP 2018

\(^2\) R. Smirnov FST 2017
Focus on wall response induced H recycling and retention from tungsten in ITER relevant conditions:

- W divertor temperature from 500K to 1400K
- W divertor exposed to DT flux $\nu H, i_n \approx 10^{24} m^{-2} s^{-1}$
- High recycling regime $\Gamma_{H,in} \approx \Gamma_{H,out}$

H molecular recombination and desorption from W:
- Second order kinetic: $\alpha = 2$
- Large recombination and reabsorption barrier $E_r \approx 1.4eV$ $^1$, $E_{S\rightarrow b} = 2eV$ $^2$

H transport and trapping in W:
- Diffusion $E_D \approx 0.2eV$ $^3$ and vacancy and defects with $E_{dt} \approx 0.85 - 2eV$ $^4$
- Implantation of He in W can strongly affect H transport and recycling in W $^5$

Modeling of tungsten wall response in ITER relevant conditions

Continuum models of hydrogen retention and recycling - and thus of wall response - include hydrogen transport, desorption and trapping in material in various regimes

**Reaction-diffusion equations**

**Bulk:**

$$\frac{\partial c_{H,\text{trap}}}{\partial T} = D \frac{\partial^2 c_H}{\partial x^2} - \frac{\partial c_{H,\text{trap}}}{\partial T} + \frac{\Gamma_{H,in}}{\lambda_{\text{imp}}},$$

**Surface:**

$$\frac{\partial c_S}{\partial T} = -K_r c_S + K_{b\rightarrow s} c_{H,0} - K_{s\rightarrow b} c_s.$$

$^2$ D. Johnson J. Mater. Res. 2010
$^3$ N.Fernandez Acta Materialia 2015
$^4$ E. Hodille Nuclear Fusion 2017
$^5$ M.J. Baldwin Nuclear Fusion 2017
Influence of surface processes on hydrogen outgassing from W remains uncertain in ITER relevant conditions

- Physical parameters of continuum models can be empirically constrained with dedicated controlled experiments (TDS, permeation,...) but large uncertainties might remain. For instance, some H surface processes on W still not well understood:
  - $H_2$ dissociation rate on W independent of H pressure \(^1\)
  - Decrease of activation energy of H desorption when W surface is saturated with H \(^2\)
- Such processes may be important in fusion relevant outgassing regimes!
- Within this framework, dynamic wall outgassing response is analyzed here using
  1. R-D models (FACE) to characterization of bulk and surface responses in relevant ITER operational regime

\[
\begin{align*}
\Gamma_{H,in} & \rightarrow \text{bulk} \\
\Gamma_{b \rightarrow s} & \rightarrow \text{surface} \\
\Gamma_{s \rightarrow b} & \rightarrow \text{bulk} \\
\Gamma_{H,\text{out}} & 
\end{align*}
\]

2. First-principle atomic modeling (LAMMPS) of H desorption from W material

\(^1\) W. Zheng, Surface Science 2006 \(^2\) P. Alnot, Surface Science 1989
Bulk outgassing response to fluctuations of plasma particle flux by diffusion of free hydrogen in the bulk is equivalent to a low-pass filter.

- Outgassing response to fluctuations of incoming particle flux $\Gamma_{H,in} = \bar{\Gamma}_{H,in} + \Delta \Gamma_{H,in} \sin \omega t$ from bulk material at fixed material temperature $T$ without traps (free H only).

- Hydrogen release from bulk governed by three time scales: $\tau_D = \frac{\lambda^2_{imp}}{D}$; $\tau_{b\rightarrow s} = \frac{\lambda_{imp}}{K_{b\rightarrow s}}$; $\tau_{bulk} = \frac{L_{bulk}^2}{D}$.

- Hydrogen release bulk onto surface assumed to be not limited by surface processes ($\frac{\tau_{b\rightarrow s}}{\tau_D} \ll 1$).

- Bulk response to $\Gamma_{H,in}$ fluctuations $\sim$ low-pass filter with $\omega_{cutoff} = \omega_D = \frac{D}{\lambda^2_{imp}}$.

- $\omega_D > 1$MHz for $400K < T$ with $\lambda_{imp} \sim 50$nm.

- Permeation barrier in material (e.g. formation of sub-surface He bubbles) may slightly affect bulk response to $\Gamma_{H,in}$ fluctuations.
Bulk outgassing response to fluctuations of plasma particle flux weakly affected by the presence of traps in material

- Outgassing response to fluctuations of incoming particle flux $\Gamma_{H,in} = \Gamma_{H,in} + \Delta\Gamma_{H,in} \sin \omega t$ from bulk material at fixed material temperature $T$ with traps

- Solute H concentration $c_{H,bulk} \sim \frac{\Gamma_{in}\lambda_{imp}}{D}$

- Effects of trapped hydrogen on dynamic outgassing only significant when trap concentration is large $c_{trap} > c_{H,bulk}$

- However, large trap concentration leads to fast hydrogen trapping ($\nu_{tr}c_{trap} \gg \nu_{dt}$) at $T < 1500K$

$\Rightarrow$ empty traps concentrations small compared to solute hydrogen concentration in material

$\Rightarrow$ Outgassing response to $\Gamma_{H,in}$ fluctuations from bulk weakly affected by hydrogen trapping in material bulk
Bulk outgassing response to fluctuations of material temperature by diffusion of free hydrogen in the bulk is equivalent to a high-pass filter

- Outgassing response to **fluctuations of material temperature** \( T = \bar{T} + \Delta T \sin \omega t \) from bulk material at fixed incoming particle flux \( \Gamma_{H,\text{in}} \) without traps (free H only)

- Hydrogen release bulk onto surface assumed to be not limited by surface processes \( \left( \frac{\tau_{b \to s}}{\tau_D} \ll 1 \right) \)

- **Bulk response to** \( T \) **fluctuations** \( \sim \) **high-pass filter** with \( \omega_{\text{cutoff}} = \omega_D = \frac{D}{\lambda_{\text{imp}}^2} \) and \( \left| \frac{\Delta \Gamma_{b \to s}}{\Gamma_{H,\text{in}}} \right| \propto \left| \frac{\Delta D}{D} \right| \approx \frac{E_D \Delta T}{T \bar{T}} \)
  - \( \omega_D > 1 \text{MHz} \) for \( 400 \text{K} < T \) with \( \lambda_{\text{imp}} \approx 50 \text{nm} \)
  - Bulk response \( \rightarrow 0 \) at high temperature

- Permeation barrier in material (e.g. formation of sub-surface He bubbles) may strongly affect bulk response to material temperature fluctuations
Surface outgassing response to fluctuations of plasma particle flux is equivalent to a low-pass filter and is affected by surface saturation with hydrogen.

- Outgassing response to fluctuations of incoming particle flux \( \Gamma_{\text{H,in}} = \Gamma_{\text{H,in}} + \Delta \Gamma_{\text{H,in}} \sin \omega t \) from material surface at fixed material temperature \( T \).

- H surface concentration described by

\[
\frac{\partial c_s}{\partial t} = -K_{r,0}e^{-\frac{E_r}{T}}(c_s)^\alpha + \Gamma_{b\rightarrow s} \left( \alpha = 2 \text{ (non-saturated surf.)} \right) \left( \alpha \rightarrow 1 \text{ (saturated surf.)} \right)
\]

- Surface (linear) response to \( \Gamma_{\text{H,in}} \) fluctuations ~ low-pass filter with \( \omega_{\text{cutoff}} = \omega_R = K_F^{\alpha^{-1}} (\Gamma_{b\rightarrow s})^{1-\alpha^{-1}} \).

- Saturation of surface (\( E_r = 1.4 \text{eV} \rightarrow 1 \text{eV} \)) strongly modify characteristic frequency of surface response.
Surface outgassing response to fluctuations of material temperature is equivalent to a high-pass filter and may be large when surface is not saturated.

- Outgassing response to fluctuations of material temperature $T = \bar{T} + \Delta T \sin \omega t$ from surface material at fixed incoming particle flux ($\Gamma_{b \rightarrow s} = \Gamma_{H,in}$).

- H surface concentration described by:
  \[
  \frac{\partial c_s}{\partial t} = -K_{r,0}e^{-E_r/T}(c_s)^{\alpha} + \Gamma_{b \rightarrow s}
  \]

- Surface response to $T$ fluctuations $\sim$ high-pass filter with $\omega_{cutoff} = \omega_{R} = K_{r}^{\alpha^{-1}}(\Gamma_{b \rightarrow s})^{1-\alpha^{-1}}$ and $\frac{|\Delta \Gamma_{H,out}|}{\Gamma_{H,out}} \propto \frac{E_r \Delta T}{T \Gamma_{H,permeation}}$

- Small fluctuations of the material surface temperature can induce large outgassing response at high frequency when surface is non-saturated.

\[
\hat{\omega} = \omega / \omega_{R}
\]
Fluctuations of temperature near material surface may be large due to reduced thermal conductivity caused by impurities and defects.

- H outgassing material response to temperature fluctuations governed by activated processes:
  \[
  \frac{\dot{\Gamma}_{\text{H, out}}}{\Gamma_{\text{H, out}}} \propto \frac{E}{T^2} \Delta T \frac{\omega/\omega_c}{\sqrt{(\omega/\omega_c)^2 + 1}}
  \]

- Rudimentary model of heat deposition on PFCs surface shows that
  \[
  \frac{\tilde{\Gamma}}{\tilde{T}} \propto \frac{\kappa}{\rho c_p} \frac{\bar{Q}}{\bar{Q}} \frac{1}{\sqrt{(\omega/\omega_Q)^2 + 1}} \quad \text{with} \quad \omega_Q \approx \frac{L_{\text{bulk}}}{\kappa}
  \]

- Modeling of realistic dynamic temperature response of PFCs is complex and must include
  - Realistic PFC geometry and cooling components
  - Reduction of thermal conductivity due to impurities and defects in material\(^1,2\), which may lead to larger temperature near the surface
  - Macroscopic plasma dynamics such as strike point motion due to vertical plasma displacement\(^3\) (e.g. strike point excursion in ITER \(\sim\) several cms)

- **Realistic thermal wall model must be provided to predict outgassing material response to heat flux fluctuations … but any experimental measurements of wall temperature fluctuations in tokamak will be extremely valuable!**

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\(^1\) S. Cui et al, Journal of Nuclear Materials 2017  
\(^2\) L. Hu et al, Appl. Phys. Lett. 2017  
\(^3\) R. Pitts et al, Nuclear Fusion 2019
H surface processes on W may provide time scale separation between plasma fluctuations and outgassing response from W wall

- Outgassing response from wall to fluctuations of plasma particle flux $\sim$ low-pass filter
- Outgassing response from wall to fluctuations of material temperature $\sim$ high-pass filter
- Outgassing response from W bulk determined by free H diffusion ($\omega_{\text{cutoff}} \approx \omega_D = \frac{D}{\lambda_{\text{imp}}^2}$)
- Outgassing response from W surface determined by H recombination and desorption ($\omega_{\text{cutoff}} \approx \omega_R = \frac{1}{T} \left( \frac{\Gamma_{b \rightarrow s}}{\Gamma_{s \rightarrow b}} \right)^{1-\alpha}$)
- Modeling of thermal fluctuations induced by fluctuations of heat flux difficult because of uncertainties in effects of implantation of plasma species on thermal conductivity of W
- In summary, H surface processes on W may induce time scale separation between slow wall outgassing response and fast plasma fluctuations observed in divertor:
  - $\omega \neq \omega_{\text{cutoff}}$: may largely simplified coupling of SOL turbulence simulations to wall dynamics
  - $\omega \approx \omega_{\text{cutoff}}$: dephasing between neutral outgassing flux and plasma flux

de \Rightarrow \text{Characterization of H processes on W surface with MD simulations}
Can H desorption from W be modeled with currently available W-H interatomic potentials?

- First-principle atomic modeling (MD simulations) may help to identify and characterize key mechanisms governing H outgassing from W:
  - Thermal desorption
  - Ion-induced desorption
  - H surface saturation effects

- Two types of W-H interatomic potentials available in literature: Tersoff \(^1\) & EAM \(^2\) potentials
- H molecular desorption from W cannot be reproduced with Tersoff potential \(^3\)

**But can EAM interatomic potential model H desorption from W?**

- Modeling of H desorption from W with MD simulations (LAMMPS):
  - Kinetic of surface desorption a priori unknown:
    \[
    \Gamma_{\text{desorption}} = K_0 e^{-\frac{E_{\text{des}}}{T} c_s} c(c_H, T)
    \]
  - MD simulations method to characterize **non-perturbed** H desorption at various material temperature T and various total amount of H in simulation to obtain \(\Gamma_{\text{desorption}}(c_s, T)\) as a function of the H surface concentration \(c_s\)
  - Kinetic order of desorption \(\alpha\) and activation energy of desorption \(E_{\text{des}}\) can be derived from \(\Gamma_{\text{desorption}}(c_s, T)\), provided that H transport \(\tau_D = \frac{L^2}{D} \) is faster than H desorption \(\tau_{\text{des}} = K_T c_s^{\alpha-1} \Rightarrow L \sim 15\text{nm with } T = 900K - 1400K

1. N. Juslin JAP 2005
2. L. Wang JPCM 2017
3. J. Guterl JNM 2013
Hydrogen molecular desorption from W simulated with EAM potential at low H surface concentration in good agreement with experimental data

- MD simulations with EAM potential show H desorption from W as H$_2$ between $T=900$-$1500$K at low H surface concentration ($c_s\lambda_0^2 << 1$):
  $$\Gamma_{\text{desorption}} = K_0 e^{-\frac{E_{\text{des}}}{T}} c_s(\lambda)$$  with $\alpha = 2$

- Activation energy for molecular desorption from W $<100>$ surface $E_{\text{des}} \sim 1.4 - 1.6$eV **in good agreement with experimental H$_2$ desorption activation energies**:
  - Markelj$^1$ (2013): 1.6-1.7eV up to 2.2eV (polycrystalline)
  - Tamm$^2$ (1969): single crystal $<100>$: 1.4eV

- Pre-exponential factor $K_0 \sim v_0^2\lambda_0^2 \sim 10^{-6}$m$^2$s in good agreement with experimental estimations:
  - Markelj$^1$ (2013): $2 - 7 \times 10^{-7}$ m$^2$s
  - Tamm$^2$ (1969): single crystal $<100>$: $4 \times 10^{-6}$ m$^2$s

- EAM potential well reproduces thermal H molecular desorption from W at low H surface concentration (unlike Tersoff potential)!

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Hydrogen molecular desorption simulated with EAM potential at high H surface concentration in qualitative agreement with experimental data

- H desorption from W saturated surface \((c_s \lambda_0^2 > 0.1)\) expected for ITER relevant conditions

- H surface concentration determined by equilibrium with H bulk concentration:

\[
K_r \ c_s^\alpha = \frac{D}{L} c_{\text{bulk}}
\]

- H bulk and surface concentrations varied through total amount of H in MD simulation:

\[
N_{\text{tot}} = \int_S c_s \, dS + \int_V c_{\text{bulk}} \, dV
\]

- Narrow parameter range to simulate H desorption from W saturated surface:
  - H desorption slow \(E_r > 1 \text{eV} \Rightarrow T > 900 \text{K}, c_s > 0.1\)
  - Formation of H platelet (self-clustering) at large bulk concentration \(^2\) induces massive H trapping \(\Rightarrow c_{\text{bulk}} \lambda_0^3 < 0.04 \Rightarrow c_s < 0.5\) and \(T<1400 \text{K}\)

- Evolution of \(E_r\) with \(c_s\) at large surface concentration in qualitative agreement with experimental data

- Transition from second to first order kinetic of desorption as surface becomes saturated

\(^1\) P. Alnot Surface Science 1989
\(^2\) R. Smirnov Nuclear Fusion 2018
A “mysterious” H precursor state for molecular desorption on W in simulations...

- H desorption from W usually assumed to result from the recombination of two H atoms into a molecule, which immediately desorbs.

- However, H molecular desorption on W follows an H precursor state\(^1\) in MD simulations using the EAM potential, whether W surface is saturated with H or not:
  1. recombination of two thermalized (cold) H into molecule
  2. dissociation of newly formed molecule into one cold and one hot H atom onto W surface
  3. molecular desorption results from the recombination of the hot atom with another cold atom

- Lifetime of molecules newly formed by two cold H is very short (\(t<0.1\)ps): no contradiction a priori with \(H_2\) dissociation on W\(^1\)

- W-H EAM potential fitted using bulk processes only and uncertainties remain for H surface processes on W:
  - additional DFT simulations required to determine whether this H precursor state is an artefact from the EAM potential
Ion-induced desorption and ion-induced detrapping may affect H recycling and retention in fusion relevant conditions

- Hydrogen recycling and retention in PFCs are usually assumed to be governed only by activated (thermal) processes (e.g. diffusion, trapping/detrapping in defects, desorption, ...)
- However, large amount of hydrogen can be present near material surface (e.g. H super-saturation observed experimentally \(^1\)) and interact with large flux of impinging particles, resulting in ion-induced processes
  - Example: well-known Eley-Rideal recombination of H on W surface with low-energy impinging H \(^2\)
- Rudimentary model of ion-induced desorption and ion-induced detrapping:
  \[
  \frac{dc_{H,\text{trap}}}{dt} = -\sigma_{dt} c_{H,\text{trap}} \Gamma_{\text{in}} - v_{dt} c_{H,\text{trap}} + v_{tr} (c_{\text{trap}} - c_{H,\text{trap}}) c_{H} \\
  \frac{dc_{s}}{dt} = -\sigma_{\text{des}} c_{s} \Gamma_{\text{in}} - K_{r} c_{s}^{g} + \Gamma_{b\rightarrow s}
  \]
- Effects of ion-induced processes on H recycling are significant when
  \[
  \frac{\sigma_{dt}}{\sigma_{\text{des, crit}}} = \frac{v_{dt}}{\Gamma_{H}} \quad \text{or} \quad \frac{\sigma_{\text{des}}}{\sigma_{\text{des, crit}}} = \frac{K_{r}^{g-1}}{\Gamma_{H}^{g-1}} (1 - R)^{1-\alpha^{-1}}
  \]
  \[\Rightarrow\] Estimations of \(\sigma_{\text{des}}\) and \(\sigma_{dt}\) with MD simulations

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1. L. Gao Nucl. Fusion 2016
2. S. Markelj Applied Surface Science 2013
MD simulations suggest that ion-induced desorption may affect H recycling in various divertor plasma regimes

- MD simulations of ion-induced H desorption on W with EAM potential
- H impinging on W with 50% H coverage induces:
  - Ion-induced H molecular desorption (Eley-Rideal)
  - H adsorption/implantation
  - H reflection
  - Ion-induced H atomic desorption
- Eley-Rideal cross section: $\sigma \sim 1 - 5\text{Å}^2$ in agreement with experimental/theoretical observations: $\sigma \sim 0.5 - 1\text{Å}^2$
- Ion-induced desorption may increase:
  - H molecular desorption at low energy
  - H effective reflection at high energy
- Some experimental observations in DIII-D suggest such ion-induced desorption during ELMs
- Electronic effects may strongly affect Eley-Rideal mechanism at low energy

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2. I. Bykov PSI 2018
Benchmarking and improvement of EAM W-H interatomic potential against DFT simulations for H surface processes on W is crucial!

- EAM potential seems to able to reproduce some key features of H desorption from W (second-order thermal molecular desorption from non-saturated surface & Eley-Rideal H recombination)
- However, EAM potentials have known intrinsic limitations in modeling of BCC metal.
- Moreover, the current EAM H-W potential does not well reproduce features of H$_2$ and H-W interactions in vacuum predicted with DFT (in contrast with the Tersoff potential!)
- Is the plateau in H-H interaction energy at $r = 1\text{Å}$ obtained with the EAM potential responsible for the H recombination precursor state observed in MD simulations?
- Can EAM potential actually be used to model H-W surface processes and accommodate multi-components chemistry (SiC-H-W, W-H-N) relevant for fusion reactor conditions?
  - See poster PA069 (S. Bringuier)

1 L. Yang JNM 2018
2 Bringuier this conference PQ069
Outgassing response from wall to
- fluctuations of plasma particle flux \( \sim \) low-pass filter
- fluctuations of material temperature \( \sim \) high-pass filter

W wall response to plasma fluctuations strongly determined by surface processes (desorption, saturation), which may induce time scale separation between fast plasma fluctuations and slow outgassing response.

Atomic characterization of H surface processes on W with molecular dynamics simulations:
- Unlike bond-order potential (Tersoff) potential, recently developed EAM potential can reproduce H thermal molecular desorption from W
- MD simulation of H desorption from W surface saturated with H in qualitative agreement with experimental observations

MD simulations framework developed to characterize ion-induced desorption processes

Uncertainties remain in the modeling of H surface processes on W with EAM potential:
- Validation of the EAM W-H interatomic potential with DFT simulations of surface processes is necessary!