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UEDGE-CRUMPET predicted isotopologue effect on atomic and molecular emission in DIII-D high-recycling divertor plasmas

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ABSTRACT

UEDGE-CRUMPET simulations indicate the impact of the molecular hydrogenic isotopologue effect under highrecycling LFS divertor conditions in DIII-D to be negligible for electron density and temperature profiles at the LFS target plate. A 30% decrease in molecular content, accompanied by a 10% increase in atomic content, is predicted for deuterium compared to hydrogen. The predicted isotopologue effect on the radiative power balance, validated with calibrated spectroscopy, is found to be small despite a 20% increase in LFS divertor molecular band emission for deuterium compared to hydrogen. The predictions and measurements show a negligible contribution of molecularly-induced atomic and direct molecular emission to the total radiative power balance under high-recycling conditions, consistent with previous EDGE2D-EIRENE investigations. The UEDGE-CRUMPET simulations were performed using effective hydrogen and deuterium rates considering molecular breakup and excitation processes for H_2 and D_2 , calculated by the CRUMPET collisional-radiative model.

1. Introduction

Molecular processes affect the plasma particle, momentum, and power balances via e.g. molecularly-assisted recombination, plasma-molecule friction, and molecular radiation, respectively. The role of molecular processes is the largest for plasma electron temperatures $1~\lesssim~T_e~\lesssim 3$ eV [1], as the atomic ionization sink and electron-ion recombination source are weak in this temperature range. Thus, molecular processes may play a role in the onset of detachment [2], the prospective operating scenario for next-step fusion devices. Hence, it is important for our predictive modeling capabilities to understand the role of molecular processes on divertor conditions, including the onset of detachment.

Tokamaks with carbon-based plasma-facing components (PFCs), such as DIII-D, are well suited for investigating the role of molecules in divertor plasmas as plasma target recycling occurs primarily as molecules [3,4]. Previous EDGE2D-EIRENE [5,6] simulations [7] of the ohmic, high-recycling deuterium-fueled DIII-D plasmas in the lower-single null configuration analyzed here predicted a mismatch of electron temperature and density profiles at the low-field side (LFS) divertor target plate and overestimated the Fulcher-band $(d^3\Pi_{\mu} \rightarrow a^3\Sigma_{\pi}^+)$

emission by a factor of 6 compared to measurements. The EDGE2D-EIRENE predictions were performed using hydrogen molecular data available in AMJUEL [8] with internal re-scaling of the molecular reaction rates by EIRENE to correspond to deuterium rates.

This work uses the edge-fluid code UEDGE [9] coupled to the CRUMPET collisional-radiative (CR) model [10] to assess whether molecular isotope (isotopologue) effects can explain the observed codeexperiment disagreement in Ref. [7] and compares the UEDGE-CRUM-PET predictions to those in Ref. [7]. The isotopologue effect on plasma target conditions and atomic and molecular emission is evaluated by comparing UEDGE-CRUMPET simulations using isotopologue-dependent rates and reactions in the CRUMPET CR model, as explained in Section 3.1. UEDGE-CRUMPET predictions are post-processed using AMJUEL to compare the role of atomic and molecular data for identical plasma conditions. The predictions are compared to DIII-D measurements in deuterium plasmas to assess the quantitative codeexperiment agreement of UEDGE-CRUMPET and the role of direct molecular emission (due to radiative transitions between molecular electronic states) and molecularly-induced atomic emission (due to production of electronically excited atoms from molecular breakup

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chains, which undergo subsequent radiative transitions) on the radiative power balance. The DIII-D measurements in Ref. [7] have been amended by measurements of the recently installed high-resolution vacuum ultraviolet (HR-VUV) spectrometer [11] to capture and resolve the Lyman $(B^{1}\Sigma_{u}^{+} \rightarrow X^{1}\Sigma_{g}^{+})$ and Werner $(C^{1}\Pi_{u} \rightarrow X^{1}\Sigma_{g}^{+})$ band emission in the vacuum-UV range.

2. Experimental plasmas

This work analyzes ohmic deuterium-fueled plasmas in DIII-D under high-recycling conditions at the LFS divertor target. The plasmas analyzed belong to the series of discharges described in detail in Ref. [7] and are chosen as high quality and extensive diagnostic data is available. The plasmas are supplied 0.9–1.1 MW ohmic heating power with 1.1 MA plasma current and 2.1 T toroidal field strength. The ion **B** × ∇ B-drift direction is into the lower divertor. Chargeexchange ion-temperature measurements are performed using 10-ms neutral beam blips in 100-ms periods. Intrinsic carbon, sputtered from the ATJ graphite divertor PFCs, is the main plasma impurity species.

The radial electron density and temperature profiles at the LFS midplane (n_e^{LFS-mp}) and T_e^{LFS-mp} , respectively) were measured using Thomson Scattering (Fig. 1a,b) and the radial location of the separatrix determined by the extended two-point model relationship outlined in Ref. [12]. The electron density and temperature radial profiles at the LFS target plate (n_e^{LFS-t}) and T_e^{LFS-t} , respectively) were measured by the Divertor Thomson Scattering (DTS) [13] viewing chord closest to the target plate, centered approximately 5 mm vertically above the target plate (Fig. 2a,b). The radial ion saturation current at the LFS target plate (j_{sat}^{LFS-t}) profiles were measured using Langmuir probes (Fig. 2c).

The radial profiles of the 656.1 nm Balmer- α (n=3 \rightarrow n=2) and 121.6 nm Lyman- α (n=2 \rightarrow n=1) line emission were measured using the Multichordal Divertor Spectrometer (MDS) [14] and Divertor Survey, Poor Resolution, Extended spectrometer (DivSPRED) [15] systems, respectively. The MDS chord viewing the LFS strike point area, covering 7.4 nm of spectral range when centered at 600 nm, was used to capture the spectral range 598–633 nm, covering the D₂ $d^3\Pi_{\mu} \rightarrow a^3\Sigma_{\alpha}^+$ Qbranch up to the fourth vibrational quantum states in four repeat plasma discharges [7]. The dispersion of the MDS spectrometer is $7.3 \times$ 10^{-3} nm/pixel, yielding a resolving power of 82,000 at 600 nm, which is sufficient to resolve the molecular Fulcher-band transitions. The total Fulcher-band emission was determined by fitting a vibrational temperature to the vibrational distribution of the measured bands, similar to Ref. [16]. The newly installed HR-VUV spectrometer with sub-ångström optical resolution was used to measure the emission in the spectral range of the Lyman $(B^1\Sigma_u^+ \rightarrow X^1\Sigma_g^+)$ and Werner $(C^1\Pi_u \rightarrow X^1\Sigma_g^+)$ band emission in the 90-150 nm spectral range along the DivSPRED line-of-sight.

3. Setup of UEDGE-CRUMPET simulations

The multi-fluid code UEDGE [9] is used to simulate the scrape-off layer (SOL) plasma and predict neutral atomic and molecular density, momentum, and temperature for conditions in the divertor. UEDGE solves the Braginskii fluid equations in the parallel-**B** direction, and a purely diffusive, anomalous radial transport model is assumed in this work. Parallel plasma transport is taken to be classical, with imposed flux limits approximating kinetic effects and preventing excessive thermal transport driven by temperature gradients. The simulations assume 1.1 MW of power entering over the core boundary, distributed equally between ions and electrons, and prescribe a constant plasma density of 3.4×10^{19} m⁻³ at the core boundary of the computational domain.

Radially varying particle and thermal diffusivities, chosen to match the measured radial electron temperature and density profiles at the LFS midplane and target plate as closely as possible within the uncertainty of the measurements, are prescribed vertically above the



Fig. 1. Measured electron temperature (a) and density (b) radial profiles at the LFSmp. UEDGE-CRUMPET predicted profiles using hydrogen (red) and deuterium (blue) effective molecular rates are shown and the separatrix location is marked by a vertical dashed line. The particle and thermal diffusion coefficient radial profiles at the LFS-mp, applied vertically above the X-point, are also presented (c). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

X-point (Fig. 1). A transport barrier is imposed by reducing the electron thermal and particle diffusivities over the separatrix (Fig. 1c). As ion upstream profile measurements are not available, a spatially constant ion thermal diffusivity ($\chi_{i,\perp} = 0.75 \text{ m}^2 \text{ s}^{-1}$) is assumed throughout the numerical domain. The resulting UEDGE-predicted LFS midplane electron temperature and density is $T_e^{LFS-mp} = 46 \text{ eV}$ and $n_e^{LFS-mp} = 2.0 \times 10^{19} \text{ m}^{-3}$, respectively. The predicted T_e^{LFS-mp} is 7 eV higher than the EDGE2D-EIRENE simulations presented in Ref. [7], which is expected to affect the predicted target conditions compared to Ref. [7].

In the divertor, vertically below the X-point, radially constant particle and thermal electron diffusivities are assigned (initially $D_{\perp} =$ $1.5 \text{ m}^2 \text{ s}^{-1}$, $\chi_{e,\perp} = 1.5 \text{ m}^2 \text{ s}^{-1}$, and $\chi_{i,\perp} = 0.75 \text{ m}^2 \text{ s}^{-1}$) as there is no transport barrier between the SOL and the PFR. The target profiles were found to be robust to changes in the radial divertor diffusivities: varying the diffusivities over two orders of magnitude resulted in a factor of 3 change in peak electron density and temperature at the LFS target plate. Instead, the target profiles show a strong dependence on the upstream radial transport coefficients and, thus, the LFS-mp profiles.

The transport of deuterium ions and intrinsic carbon impurity ions is simulated, including $\mathbf{E} \times \mathbf{B}$ and $\mathbf{B} \times \nabla \mathbf{B}$ drift flows. Deuterium atoms are simulated by an inertial neutral model, describing perpendicular transport by charge-exchange diffusion and solving a full momentum equation in the parallel-B direction. All plasma fluxes impinging on the targets are assumed to recycle as thermal molecules and the atom source is volumetric dissociation of molecules and ion-electron recombination. Diffusive transport of the recycled molecules is assumed. Physical and chemical sputtering of carbon as atoms at the divertor targets is considered using the 1997 Haasz–Davis sputtering yield [17]. The one-sided Maxwellian flux of atoms and molecules impinging on the poloidal and radial domain boundaries are uniformly removed at 1% rate, approximating particle removal by the cryopumps.

The UEDGE molecular model [18] has been extended to consider molecularly-induced plasma particle and energy sinks and sources [19] by coupling to the CR model CRUMPET [10]. CRUMPET calculates the

effective particle and energy sinks and sources due to molecular reactions associated with the time-dependently resolved electronic ground state atoms and electronic and vibrational ground state molecules in UEDGE. The effective rates are calculated using an effective rate matrix, considering transitions via intermediate states with short life times, as outlined in Refs. [19,20]. The particle and energy sinks and sources considered include e.g. molecularly-assisted recombination, binding energy losses, direct molecular emission, and molecularly-induced atomic emission, but does not yet consider plasma momentum losses due to ion–molecule friction.

The UEDGE-CRUMPET predictions were subsequently post-processed by CRUMPET to recover the populations of the electronically and vibrationally excited states, which are not resolved in UEDGE, to evaluate the UEDGE-CRUMPET predicted atomic line and molecular band emission. The atomic line and molecular band emission were calculated locally for each cell by multiplying the atomic and (vibrationally unresolved) molecular upper excited electronic state populations with calculated Einstein coefficients. A synthetic spectroscopy module was used to determine the emission profiles as measured by the synthetic spectroscopy systems. To isolate the role of the atomic and molecular data and the CRUMPET CR model from differences in the plasma predictions, the corresponding AMJUEL [8] rates were evaluated using the same synthetic spectroscopy module as the CRUMPET results. The AMJUEL post-processing is applied to the UEDGE-CRUMPET H₂ simulations, as the AMJUEL data is given for hydrogen, using the same reactions as in Ref. [7] (see Appendix A.3 for details).

3.1. Molecular rates and data

Two sets of UEDGE simulations employing CRUMPET molecular rates for hydrogen and deuterium molecules, respectively, were performed to assess the isotopologue effect on the divertor plasma conditions in DIII-D under high-recycling LFS target and detached HFS target conditions. The UEDGE simulations evaluate the transport of deuterium in both cases, to isolate the effect of molecular isotopologue effects from plasma transport effects caused by the different mass of the isotopes.

The CR model applied considers a vibrationally resolved molecular electronic $X^{1}\Sigma_{g}^{+}$ ground state, vibrationally unresolved n = 2 singlet $(B^{1}\Sigma_{u}^{+}, C^{1}\Pi_{u}, EF^{1}\Sigma_{g}^{+})$ and triplet $(a^{3}\Sigma_{g}^{+}, c^{3}\Pi_{u})$ states, a vibrationally unresolved subset of n = 3 singlet $(B'^{1}\Sigma_{u}^{+} \text{ and } D^{1}\Pi_{u})$ and triplet $(e^{3}\Sigma_{u}^{+})$ and $d^{3}\Pi_{u}$ states, and the eight first electronic states of the hydrogen atom. Transitions into the dissociative triplet $b^{3}\Sigma_{u}^{+}$ state are included and assumed to immediately dissociate into two electronic ground-state atoms. The H₂ CR model includes reactions involving both H⁻ and H₂⁺, whereas the D₂ CR model omits the dissociative attachment (D⁻) channel as it is assumed negligible for D₂ [21,22].

The CR model utilizes reaction data from the EIRENE databases AMJUEL, H2VIBR [23], and HYDHEL [24]. Electron-impact electronic transition and dissociation cross-sections are taken from the molecular convergent close-coupling (MCCC) database [25] and the molecular radiative transition coefficients are calculated using Ref. [26]. MCCC rates are not available for the n = 3 molecular electronic states: instead, corresponding rates for the n = 2 state closest in energy were used and the threshold of the ionization cross-sections shifted to correspond to the ionization potential of the molecular n = 3 states. The D₂ CR model scales the vibrationally resolved H2VIBR rates for H2 according to the potential energy of the corresponding D₂ states according to Ref. [27], Ch. 7. The electron impact cross-sections for the $D_2 X^1 \Sigma_{\alpha}^+$ -state are available in the MCCC database and used: for other electron-impact transitions the MCCC database H₂ rates are applied. A detailed description of the applied CR models for H_2 and D_2 is available in the appendix.



Fig. 2. Radial profiles of the electron temperature (a) and density (b) at the LFS divertor target plate as a function of the radial distance from the separatrix at the LFS midplane measured by DTS. The ion saturation current as a function of the radial distance from the separatrix at the LFS target measured by Langmuir probes is also shown (c). UEDGE-CRUMPET predicted profiles using hydrogen (red) and deuterium (blue) effective molecular rates are shown and the separatrix location is marked by a vertical dashed line. The cell centers of the UEDGE grid are marked by crosses. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

4. Results

The UEDGE-CRUMPET predictions indicate the isotopologue effect has a negligible impact on the T_{e}^{LFS-t} , n_{e}^{LFS-t} , and j_{sqt}^{LFS-t} radial profiles (Fig. 2a-c). However, UEDGE-CRUMPET predicts 30% lower molecular content in the simulations using effective D₂ rates compared to the simulations using effective H₂ rates, accompanied by a 10% increase in atomic content due to differences in the effective dissociation rates of H₂ and D₂. The atomic content in the numerical domain is a factor of 10 and 15 higher than the molecular content for the hydrogen and deuterium simulations, respectively. The decrease in molecular content for D₂ compared to H₂ occurs predominantly in the far-SOL, poloidally upstream of the target, where the plasma densities and temperatures are low. Consequently, the difference in atomic and molecular content between the hydrogen and deuterium UEDGE-CRUMPET simulations does not impact the plasma target conditions. The difference in molecular content is mediated by an increase in the ion-conversion (H⁺ + H₂ \rightarrow H + H₂⁺) rate of D₂ compared to H₂ in the applied CR model (see appendix). The ion-conversion rate limits dissociation via reaction chains for $T_e > 3$ eV, explaining the decrease in molecular content for D₂ compared to H₂.

The peak of the UEDGE-CRUMPET predicted T_e^{LFS-t} -profiles occurs at the same radial location as the DTS measurements, and the predicted profiles lie within the uncertainty of the measurements radially outside the T_e^{LFS-t} -peak (Fig. 2a). The predicted radial temperature gradients in the private-flux region (PFR) are smaller than for the measurements, resulting in an overestimation of T_e^{LFS-t} by $\sim 1 \text{ eV}$ in the PFR. The UEDGE-CRUMPET predicted radial T_e^{LFS-t} -profiles are in better agreement with the measurements compared to the EDGE2D-EIRENE predictions in Ref. [7], indicating different divertor target conditions in the two codes.

The n_e^{LFS-t} -predictions lie within the uncertainty of the measurements for most of the radial profile, but do not capture the strongly peaked density at the separatrix observed for the DTS measurements, resulting in an underestimation of the peak n_e^{LFS-t} by a factor of ~4 (Fig. 2b). A similar underestimation of the peak n_e^{LFS-t} was observed in Ref. [7] and is further elaborated in Ref. [28]. It is possible that the anomalous, diffusive radial transport model cannot support the strong radial density gradients measured by the DTS system. Investigation of the mismatch is deferred to future publications. The underestimation of peak n_e^{LFS-t} by UEDGE-CRUMPET results in a corresponding underestimation of peak j_{sat} by ~50%, consistent with the EDGE2D-EIRENE predictions in Ref. [7].

The synthetic DivSPRED radial Lyman- α (n=2 \rightarrow n=1) line emission profiles along the LFS target plate, obtained by post-processing the UEDGE-CRUMPET simulations, mostly lie within the indicative 25% uncertainties of the spectroscopic measurements (Fig. 3). For $r^{LFS-t} - r^{LFS-t}_{sep} < 8$ mm, the n=2 \rightarrow n=1 emission predictions are poorest and the emission peak is underestimated by ~30% compared to the experimental measurements. This underestimation correlates well with the overestimation of T_e^{LFS-t} and underestimation of n_e^{LFS-t} for $r^{LFS-mp} - r^{LFS-mp}_{sep} < 1.2$ mm by UEDGE-CRUMPET, with the experimentally observed n=2 \rightarrow n=1 peak corresponding to the experimentally observed n=2 \rightarrow n=1 peak corresponding to the experimentally observed peak of n_e^{LFS-t} (Fig. 2a–b). The UEDGE-CRUMPET predicted population of the n = 3 electronically excited state due to molecular processes contributes 2% to the total n=2 \rightarrow n=1 emission, indicating the role of molecularly-induced n=2 \rightarrow n=1 emission to be negligible under high-recycling conditions.

The UEDGE-CRUMPET predicted peak n=2 \rightarrow n=1 line emission is within 5% of the AMJUEL predictions and in good agreement with the EDGE2D-EIRENE predictions in Ref. [7] despite the observed difference of the T_e^{LFS-t} -profiles (Fig. 3). This is consistent with the n=2 \rightarrow n=1 emission being strongest close to the ionization front (as the electron temperatures are sufficient for excitation of the bound electrons), where the EDGE2D-EIRENE predictions are in good agreement with measurements as discussed in Ref. [28]. Note that re-calibration



Fig. 3. DivSPRED measured (circles) and UEDGE-CRUMPET predicted LFS divertor $n=2\rightarrow n=1$ emission as a function of radial distance from the separatrix at the LFS target plate using hydrogen (red) and deuterium (blue) effective molecular rates. Indicative uncertainties of 25% for the experimental measurements and the AMJUEL-predicted $n=2\rightarrow n=1$ emission are marked. The separatrix is marked by a vertical dashed line. Note that the DivSPRED data has been re-calibrated compared to the measurements presented in Ref. [7]. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

of the DivSPRED data has been performed, increasing the measured $n=2\rightarrow n=1$ line emission by up to 40% compared to the corresponding measurements presented in Ref. [7].

The radial Balmer- α (n=3 \rightarrow n=2) line emission profiles of the synthetic MDS system are within the uncertainty of the measurements in the high-recycling LFS divertor but underestimates the peak $n=3\rightarrow n=2$ line emission for the detached HFS divertor by a factor of ~7 (Fig. 4a). UEDGE-CRUMPET predicts 80% of the total $n=3\rightarrow n=2$ line emission in the HFS divertor to be due to molecularly-induced $n=3\rightarrow n=2$ emission, similar to AMJUEL predictions and the EDGE2D-EIRENE predictions in Ref. [7]. This is indicative of $n=3\rightarrow n=2$ emission to be mainly due to molecularly-induced atomic emission under detached conditions. The molecular rates and, consequently, the molecularly-induced atomic emission are strongly dependent on the vibrational distribution of the molecular electronic ground state [1]. Thus, surface processes, such as the Eley-Rideal and the Langmuir-Hinshelwood mechanisms, may result in release of molecules with non-thermal vibrational distribution from the target plates [29], as has been observed for graphite targets [4]. Such deviations from the assumed recycling of molecules at their electronic and vibrational ground-state may explain the underestimation of $n=3\rightarrow n=2$ emission at the HFS. Underestimation of electron-ion recombination into the n = 3 state, which is sensitive to the local plasma density and temperature, in UEDGE-CRUMPET could also contribute to the observed underestimation in $n=3 \rightarrow n=2$ emission. Despite UEDGE-CRUMPET predicting 28% lower molecular content using D₂ effective rates compared to when using H₂ effective rates, the molecularly-induced $n=3\rightarrow n=2$ emission in the HFS divertor is unaffected. This is consistent with the increase in molecular content occurring predominantly in the far-SOL poloidally upstream of the target, where the plasma temperature is insufficient provide sufficient energy to produce n = 3 atoms from the molecular dissociation chain.

UEDGE-CRUMPET overestimates the Fulcher-band $(d^3\Pi_u \rightarrow a^3\Sigma_g^+)$ emission by a factor of 10 compared to measurements, comparable to



Fig. 4. Radial profiles of the Balmer- α (n=3 \rightarrow n=2) line (a), integrated Fulcher ($d^3\Pi_u \rightarrow a^3\Sigma_g^+$) band (b) and integrated Lyman–Werner ($B^1\Sigma_u^+ \rightarrow X^1\Sigma_g^+$ and $C^1\Pi_u \rightarrow X^1\Sigma_g^+$) band emission (c) as a function of the synthetic MDS angle normalized to the LFS strike-point location predicted by UEDGE-CRUMPET using hydrogen (red) and deuterium (blue) effective molecular rates. MDS measurements (circles) of the n=3 \rightarrow n=2 (with 25% indicative uncertainties) and $d^3\Pi_u \rightarrow a^3\Sigma_g^+$ emission are shown in (a) and (b). Corresponding AMJUEL-predicted emission (gray) and contributions from direct atomic excitation (dotted lines) are also shown. The insert in (c) shows the synthetic MDS and DivSPRED chords. The UEDGE-CRUMPET predicted ratio of molecular band emission to atomic line emission as a function of the synthetic MDS angle normalized to the LFS strike-point location is shown in (d). Note the logarithmic ordinate in (d) and that the predictions in (b) are scaled by a factor of 10 for brevity of presentation. The LFS and HFS strike-points (0° and -8.2° , respectively) are marked by dashed vertical lines. (For interpretation of this article.)

the EDGE2D-EIRENE simulations in Ref. [7] (Fig. 4b). As the excitation potential of the $d^3\Pi_u$ state is higher than the atomic ionization energy, $d^3\Pi_u \rightarrow a^3\Sigma_g^+$ emission will occur predominantly upstream of the ionization front where predictions are in good agreement with measurements [28]. Therefore, the overestimation is likely due to the applied molecular CR model rather than the plasma solution.

The $d^3\Pi_u \rightarrow a^3\Sigma_g^+$ emission is determined by the steady-state population of the $d^3\Pi_u$ state, which is predicted by CRUMPET postprocessing of the plasma solution. The $d^3\Pi_{\mu}$ population depends on the population and depopulation reactions of the state, and CRUMPET indicates the $d^3\Pi_{\mu}$ state is predominantly populated by excitation from the $X^1\Sigma_{\rm g}^+$ state. Thus, the $d^3\Pi_{\rm u} \rightarrow a^3\Sigma_{\rm g}^+$ emission depends on the vibrational distribution of the electronic ground state molecules, which is affected by the potential release of molecules with a non-thermal vibrational distribution from the target plates due to surface processes as discussed above. The fraction of molecular recycling also impacts the $d^3\Pi_u \rightarrow a^3\Sigma_{\sigma}^+$ emission: this effect is, however, expected to be small as up to 90% of the target fluxes are recycled as molecules [3]. The $d^3\Pi_{\rm u}$ state depopulation reactions are not available from Ref. [25] and are approximated as explained in Section 3.1 and the appendix, which directly affects the CRUMPET-predicted $d^3\Pi_u \rightarrow a^3\Sigma_{\sigma}^+$ emission. UEDGE-CRUMPET predicts 40% and 25% higher $d^3\Pi_{\rm u} \rightarrow a^3 \tilde{\Sigma}_{\sigma}^+$ emission than AMJUEL for H₂ and D₂, respectively, a difference that is roughly of the same order of magnitude as the uncertainty of the experimental measurements. It was not possible to compare the difference of the individual rates used CRUMPET and AMJUEL, since the AMJUEL data is given as a double polynomial fit of cumulative reactions.

UEDGE-CRUMPET predicts the Lyman–Werner band $(B^1\Sigma_{u}^+ \rightarrow X^1\Sigma_{\sigma}^+)$ and $C^1\Pi_{\mu} \rightarrow X^1\Sigma^+_{\alpha}$) emission to contribute 2%-4% of the total radiated power, comparable to initial HR-VUV measurements indicating a ~2% contribution of the observed $B^1\Sigma_u^+ \rightarrow X^1\Sigma_\sigma^+$ and $C^1\Pi_u \rightarrow$ $X^{1}\Sigma_{\alpha}^{+}$ transitions to total radiated power [30]. Radial profiles at the LFS target of the $B^1\Sigma_u^+ \rightarrow X^1\Sigma_g^+$ and $C^1\Pi_u \rightarrow X^1\Sigma_g^+$ transitions are not available as the bands could not be reliably measured during the strike-point sweep of the high-recycling cases analyzed here. However, Ref. [30] indicates that the $B^1\Sigma_u^+ \to X^1\Sigma_\sigma^+$ and $C^1\Pi_u \to$ $X^{1}\Sigma_{+}^{+}$ transitions are strongest in the SOL close to the separatrix, in qualitative agreement with the UEDGE-CRUMPET predictions (Fig. 4c). The predictions overestimate the Lyman-Werner band emission by up to a factor of 3 compared to AMJUEL predictions (Fig. 4c), and the AMJUEL-predicted Lyman-Werner band emission is comparable to that predicted by EDGE2D-EIRENE in Ref. [7]. The overestimation by UEDGE-CRUMPET compared to AMJUEL predictions is attributed to the MCCC population and depopulation rates of the $B^1\Sigma_{\mu}^+$ and $C^{1}\Pi_{u}$ states used in UEDGE-CRUMPET, which are different from the corresponding rates considered by AMJUEL.

UEDGE-CRUMPET predicts a ~20% increase in the molecular contribution to the total emission intensity for deuterium compared to hydrogen in the LFS divertor (Fig. 4d). The increase is due to an increase in $d^3\Pi_u \rightarrow a^3\Sigma_g^+$, $B^1\Sigma_u^+ \rightarrow X^1\Sigma_g^+$ and $C^1\Pi_u \rightarrow X^1\Sigma_g^+$ band emission for D₂ compared to H₂ (Fig. 4d), indicating an isotopologue effect in the UEDGE-CRUMPET simulations under high-recycling conditions in the LFS divertor. The UEDGE-CRUMPET predicted isotopologue effect is, however, not expected to be experimentally observable, as it is of the same order of magnitude as the experimental uncertainties. In the detached HFS, molecular emission is weak and the isotopologue effect is negligible, as the plasma temperatures are insufficient to electronically excite the molecules.

The n=2 \rightarrow n=1 atomic line and the molecular $B^{1}\Sigma_{u}^{+} \rightarrow X^{1}\Sigma_{g}^{+}$ and $C^{1}\Pi_{u} \rightarrow X^{1}\Sigma_{g}^{+}$ band emission, which have comparable wavelengths, are found to be the dominant atomic and molecular radiative power exhaust channels, respectively (Figs. 3 and 4a–c). UEDGE-CRUMPET predicts the $B^{1}\Sigma_{u}^{+} \rightarrow X^{1}\Sigma_{g}^{+}$ and $C^{1}\Pi_{u} \rightarrow X^{1}\Sigma_{g}^{+}$ emission to be 10% of the n=2 \rightarrow n=1 line emission in the radiative LFS divertor (Fig. 4d), and 2%–4% of the total radiated power in the computational domain. The

UEDGE-CRUMPET predicted $B^1\Sigma_u^+ \rightarrow X^1\Sigma_{\sigma}^+$ and $C^1\Pi_u \rightarrow X^1\Sigma_{\sigma}^+$ band emission are, however, a factor of 3 higher than the corresponding AMJUEL predictions (Fig. 4c). Additionally, release of molecules with a non-thermal vibrational distribution from the target plates due to surface processes, which are expected to contribute to the factor of 10 overestimation of $d^3\Pi_u \rightarrow a^3\Sigma_g^+$ emission compared to the measurements, may also result in overestimation of the predicted $B^1 \Sigma_{\mu}^+ \rightarrow$ $X^1\Sigma_{\rm g}^+$ and $C^1\Pi_{\rm u} \rightarrow X^1\Sigma_{\rm g}^+$ transitions. Therefore, the contribution of molecular emission to the total radiated power in the numerical domain is expected to be small for high-recycling divertor conditions in DIII-D and, furthermore, the impact of the isotopologue effect on the radiative power balance negligible. UEDGE-CRUMPET predicts the molecular binding energy, lost by the electrons to binding potential of the dissociation products upon dissociation, to account for $\sim 1\%$ of the total plasma input power. The molecular binding energy losses are not spectroscopically measurable and released to the targets upon surface recombination.

5. Summary

The molecular isotope (isotopologue) effect was evaluated by comparing UEDGE-CRUMPET simulations of ohmic, deuterium-fueled DIII-D plasmas in high-recycling LFS and detached HFS target plate conditions using effective H₂ and D₂ molecular rates, respectively. The simulations indicate a negligible isotopologue effect on the electron density, electron temperature, and ion saturation current at the LFS target plate. A 20% increase in molecular emission in the high-recycling LFS divertor is predicted for D₂ compared to H₂ despite a 30% decrease in net D₂ content compared to H₂ content. This observation is explained by the decrease in D₂ content compared to H₂ content occurring in the far-SOL upstream of the target, where the plasma temperatures are insufficient to electronically excite the molecules. For the same reason, molecular emission and the isotopologue effect in the HFS divertor is negligible. However, the UEDGE-CRUMPET predicts production of radiating atoms via molecular dissociation to account for 80% of atomic line emission in the detached HFS, which should be considered in the context of ITER operation.

UEDGE-CRUMPET predictions indicate the molecular emission, dominated by the Lyman–Werner $(B^1\Sigma_u^+ \to X^1\Sigma_\sigma^+ \text{ and } C^1\Pi_u \to X^1\Sigma_\sigma^+)$ band emission, to be less than 10% of the atomic line emission, dominated by the atomic Lyman- α (n=2 \rightarrow n=1) line, and 2%–4% of the total radiated power in the numerical domain, comparable to the measurements in Ref. [30]. However, UEDGE-CRUMPET overestimates the $B^{1}\Sigma_{u}^{+} \rightarrow X^{1}\Sigma_{g}^{+}$ and $C^{1}\Pi_{u} \rightarrow X^{1}\Sigma_{g}^{+}$ transitions by a factor of 3 compared to AMJUEL and overestimates the molecular Fulcher $(d^{3}\Pi_{u} \rightarrow a^{3}\Sigma_{g}^{+})$ band emission by a factor of 10 compared to experimental measurements. Thus, the molecular contribution to the global power balance is concluded to be insignificant, consistent with DIII-D measurements and the EDGE2D-EIRENE predictions in Ref. [7]. The overestimation of UEDGE-CRUMPET predicted molecular emission compared to measurements and AMJUEL predictions is attributed to the rates and reactions considered in the collisional-radiative CRUMPET model. The molecular convergent close-coupling rates applied in the CR model does not include depopulation rates for the $d^3 \Pi_u$ state, which were approximated using available triplet rates, and are expected to affect the predicted $d^{3}\Pi_{\mu} \rightarrow a^{3}\Sigma_{\sigma}^{+}$ emission. To assess the impact of the applied rates and determine what processes are responsible for the overestimation of the measured $d^3\Pi_u \rightarrow a^3\Sigma_g^+$ and $B^1\Sigma_u^+ \rightarrow X^1\Sigma_g^+$ and $C^1\Pi_u \rightarrow X^1\Sigma_g^+$ band emission, the CRUMPET CR model could be compared to established CR models, such as YACORA [31].

The UEDGE-CRUMPET model could be further developed to account for surface processes resulting in release of molecules with non-thermal vibrational distribution, which are expected to affect the population of the electronically excited molecular states. Extending the CRUMPET CR model to include molecular convergent close-coupling rates for electronically excited D₂ states and depopulation rates for the $d^3\Pi_u$

state, which are not presently available is also expected to affect the UEDGE-CRUMPET predictions. Vibrationally resolving the electronically excited molecular states, using the Franck–Condon factors in Ref. [26], is also expected to improve the accuracy of the UEDGE-CRUMPET model. The UEDGE-CRUMPET simulations presented here varies the radial thermal and particle diffusion coefficients at the LFS midplane to match the electron temperature and density profiles at the LFS midplane and along the LFS target plate. The set of diffusion coefficients applied are not necessarily a unique solution to match the predictions to the measurements. Additionally, the UEDGE-CRUMPET predictions depend on the radial boundary conditions applied and the physics included in the simulations, such as drifts and radial convection.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Andreas Holm, Mathias Groth reports financial support was provided by Academy of Finland. DIII-D and LLNL funding reports financial support was provided by US Department of Energy.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary material related to this article can be found online at https://doi.org/10.1016/j.nme.2022.101337.

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