A local-spectrum correlated model for radiative transfer in non-uniform gas media

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Abstract

A new local-spectrum correlated model is developed for the solution of the radiative transfer equation (RTE) in non-uniform gas media at high temperature. The spectral integration of the RTE is performed with the help of a new gas absorption spectrum distribution function termed the cumulative wavenumber. The new approach features local-spectrum correlation, rather than the global-spectrum correlation used traditionally. A local correction factor introduced to account for spatial variations in species concentrations and gas temperature is performed independently in each spectral interval under the assumption of local-spectrum correlation. Hence, the dependence of predictions on the selection of a reference temperature is nearly eliminated. The method is shown to be capable of treating non-gray particulates and boundaries. © 2002 Elsevier Science Ltd. All rights reserved.

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1. Introduction

In recent years, much attention has been focused on the solution of radiative transfer problems in high temperature gaseous media [1]. Special interest was directed towards methods of spectral integration, to the problems in mixtures of gases with scattering particles, and to the treatment of non-isothermal and non-homogeneous cases.

The weighted-sum-of gray-gases approach (WSGG) proposed in 1967 by Hottel and Sarofim [2] in the context of the zonal method, undertook a development in the methods based on the global absorption coefficient distribution functions (WSGG [3], SLW [4–6], ADF [7], ADFFG [8]). These methods, well-suited for the spectral integration in combination with the arbitrary methods of solution of radiative transfer equation (RTE), experience difficulties in the cases of gas mixtures or non-uniform media. The special assumptions or hybrid combinations with other methods are required...
in these cases [9]. The first development of the WSGG to the solution of RTE [3] was made only for uniform media with black boundaries. Spectral integration of RTE in the SLW method [5] leads to the appearance of Leibnitz terms in the equation for the gray gases which make the method to be ineffective. The situation was treated by the assumption of the ideal behavior of molecular spectra with temperature or by a simple neglect of the Leibniz terms [5]. The full-spectrum correlated-\(k\) distribution [10] was introduced to accommodate WSGG method in non-uniform media. To eliminate the physical inconsistency of the scaling in full-spectrum correlated ideal spectra, the ADFFG method uses the separation of the actual spectrum into fictitious classes that are better suited for full-spectrum scaling [8].

In the present work, which also is a part of the global approach, we introduce a distribution function—the cumulative wavenumber—which is convenient for the spectral integration and for the treatment of gas mixtures with non-gray particles and boundaries. This approach allows one to make an assumption of local-spectrum correlation in non-uniform media which is better justified than the assumption of ideal behavior of spectra.

2. Cumulative wavenumber

2.1. Definition

The distribution functions are usually defined on the basis of the high resolution molecular absorption cross-section. Molecular spectra of gases contain hundreds of thousands of lines and cannot be efficiently used directly in comprehensive radiative transfer calculations. Distribution functions contain compressed information about the spectra. Here, we introduce a distribution function—the cumulative wavenumber—which is a source of dynamic information about the molecular absorption spectra of gases. It contains the complete detailed information of the spectra. In general, the absorption spectrum can be reconstructed in its entirety from the cumulative wavenumber.

Consider the molecular gas absorption cross-section \(C_{\eta}\) which is a function of wavenumber \(\eta\). For any fixed value of absorption cross-section \(C\) and wavenumber \(\eta\), we define a cumulative wavenumber by the following relation:

\[
w(C, \eta) = \begin{cases} 
\sum_{i=1}^{L} (\beta_i - \alpha_i) + (\eta - \eta^*) & \text{if } C_\eta < C, \\
\sum_{i=1}^{L} (\beta_i - \alpha_i) & \text{if } C_\eta \geq C,
\end{cases}
\]

where wavenumber intervals \([\alpha_i, \beta_i]\) are obtained by the intersection of the absorption cross-section \(C_\eta\) with the line \(C_\eta = C\), and \(\eta^*\) is the largest wavenumber of the intersection for the case when, for the wavenumber variable \(\eta\), the absorption cross-section \(C_\eta\) is less than the value \(C\). This is illustrated in Fig. 1. It is easily seen from the definition that the cumulative wavenumber is a non-decreasing, continuous function of \(\eta\).

In Fig. 2, the cumulative wavenumber behavior of \(H_2O\) and \(CO_2\) at 1000 K is shown for the set of absorption cross-sections in the range \(10^{-25} < C < 10^{-17} \text{ cm}^2/\text{molecule}\). The upper curve
corresponds to $C = 10^{-17}$ cm$^2$/molecule, and the lower curve corresponds to $C = 10^{-25}$ cm$^2$/molecule in half-decade increments. The calculations were performed with the HITEMP spectral database [11].

2.2. Derivative and integration

According to its definition, the cumulative wavenumber $w(C, \eta)$ grows linearly with $\eta$ for those values of wavenumber $\eta$ where $C_\eta$ is less than the value $C$, and remains constant elsewhere. The extreme case of the cumulative wavenumber is a straight line when the value $C$ is greater than the entire absorption cross-section $C_\eta$. Therefore, the differentiation of Eq. (2) with respect to wavenumber $\eta$ yields:

$$\frac{\partial w(C, \eta)}{\partial \eta} = \begin{cases} 1 & \text{if } C_\eta < C, \\ 0 & \text{if } C_\eta > C. \end{cases}$$ (2)

This formula allows one to replace the integration with respect to the independent variable $\eta$ over the spectral region defined by the molecular absorption spectrum by the integration with respect to cumulative wavenumber

$$\int_{\{\eta : C_\eta < C\}} d\eta = \int_{\eta=0}^{\infty} \frac{\partial w(C, \eta)}{\partial \eta} d\eta = \int_{\eta=0}^{\infty} dw(C, \eta).$$ (3)
2.3. Dependence of cumulative wavenumber on mole fraction

The dependence of absorption-line blackbody distribution function on mole fraction in the SLW model has been derived in detail previously [12], and is extended here to cumulative wavenumber. Let $w_{C_Y}$ and $w_{YC_Y}$ be the cumulative wavenumber of the absorption cross-sections $C_Y$ and $YC_Y$, respectively, neglecting the effect of self-broadening. Here, $Y$ is the mole fraction of some species in the gas mixture. As seen in Fig. 3(a), the intersections of the cross-section $C$ with the spectrum $YC_Y$ produce the same wavenumber intervals as the intersections of cross-section $C/Y$ with the
spectrum. The cumulative wavenumber of the spectrum $YC_\eta$ at $C$ is, therefore, equal to the cumulative wavenumber of the spectrum $C_\eta$ at $C/Y$

$$w_{YC_\eta}(C, \eta) = w_{C_\eta}(C/Y, \eta).$$  \hfill (4)

If the mole fraction of the species included in a gas mixture changes, the cumulative wavenumber need not be re-calculated from the molecular spectrum. Rather, it can be determined using Eq. (4). This property is fundamental for the derivation of new methods of spectral integration for radiative transfer in the mixture of gases.

2.4. Absorption cross-section with a constant addition

Suppose that the absorption cross-section of a medium $C_\eta$ has a constant addition $C^*$ and is represented by the sum $C_\eta + C^*$ (superposition of the molecular absorption spectrum with the absorption coefficient of gray particles). Similar to the previous case, the intersections of cross-section $C$ with spectrum $C_\eta + C^*$ produce the same wavenumber intervals as the intersections of cross-section $C_\eta - C^*$ with the spectrum $C_\eta$ as shown in Fig. 3(b). This gives the property

$$w_{C_\eta + C^*}(C, \eta) = w_{C_\eta}(C - C^*, \eta).$$  \hfill (5)
Combination of this property with Eq. (4) yields a relationship for the cumulative wavenumber of the absorption cross-section $YC/DC_1 + C^*$:

$$w_{YC_1 + C^*}(C, \eta) = w_{C_1}(C/Y - C^*/Y, \eta).$$  \hspace{1cm} (6)

2.5. Gas mixtures

If $m$ is the number of gas species in a mixture, then, in the assumption of non-overlapping spectral lines for the absorption cross-section, $C_\eta = Y_1C_1 + Y_2C_2 + \cdots + Y_mC_m$, the cumulative wavenumber for the mixture may be expressed as

$$w_{C_\eta}(C, \eta) = w_1 \left( \frac{C}{Y_1}, \eta \right) + w_2 \left( \frac{C}{Y_2}, \eta \right) + \cdots + w_m \left( \frac{C}{Y_m}, \eta \right) - (m - 1)\eta.$$  \hspace{1cm} (7)

In combination with the gray spectrum associated with constant addition for the absorption cross-section $C_\eta + C^*$, the cumulative wavenumber is defined by the equation

$$w_{C_\eta + C^*}(C, \eta) = w_1 \left( \frac{C}{Y_1} - C^*, \eta_1 \right) + w_2 \left( \frac{C}{Y_2} - C^*, \eta_2 \right) + \cdots + w_m \left( \frac{C}{Y_m} - C^*, \eta_m \right) - (m - 1)\eta,$$  \hspace{1cm} (8)

where $w_i$ are the cumulative wavenumbers of individual species included in the mixture.

3. Spectral model

Consider a subdivision of the full range of absorption cross-sections into gray gases by the supplemental cross-sections $C_j; j = 1, \ldots, n$, where $n$ is the number of gray gases (Fig. 4). Consider the set $H_j$ as the union of all spectral subintervals for which the actual absorption cross-section $C_\eta$ lies between supplemental absorption cross-sections $C_j$ and $C_j - 1$, expressed as

$$H_j = \{ \eta; \ C_j - 1 \leq C_\eta \leq C_j \}, \quad j = 1, \ldots, n.$$  \hspace{1cm} (9)

We also have the subdivision of the wavenumber region into subintervals

$$A_i = [\eta_{i-1}, \eta_i], \quad i = 1, 2, \ldots.$$  \hspace{1cm} (10)

In the present model, we use the intersection of these two spectral subdivisions to define the modeling of fractional gray gas wavenumbers $D_{ij}$:

$$D_{ij} = A_i \cap H_j.$$  \hspace{1cm} (11)

The union of all fractional gray gas wavenumbers gives the entire wavenumber space. Although $A_i$ remains fixed for any location in the medium considered, $H_j$ and $D_{ij}$ are allowed to float with location depending on the local thermodynamic state of the gas. We apply this modeling to subdivide the spectral integration over wavenumber space into the integration over fractional gray gas wavenumbers $D_{ij}$. The subdivision into gray gases $H_j$ is used for simplification of the molecular line. Each gray gas is represented by a single value of absorption coefficient $\kappa_j$. The subdivision into wavenumber intervals $A_i$ is used for incorporation of continuous spectrum contributions (e.g., soot particles or aerosols) and non-gray boundaries. This spectral model also allows the assumption of local-spectrum correlation, which will now be developed.
4. Local-spectrum correlation

The cumulative wavenumber $w(C, \eta)$ is defined by the absorption cross-section $C_\eta$ which is a function of the local thermodynamic state of the gases (temperature, partial pressure, concentrations of species). In non-isothermal and non-homogeneous media, each location along an arbitrary path-length $s$ is characterized by its own thermodynamic state and, therefore, in general, the cumulative wavenumber is also a function of location $s$, $w(C, s, \eta)$.

Our assumption of local-spectrum correlation is that the difference between cumulative wavenumber at two adjacent supplemental cross-sections for each fractional gray gas is represented by the product of two functions, one of which depends on location $s$ and the other on wavenumber $\eta$:

$$w(C_j, s, \eta) - w(C_{j-1}, s, \eta) = u_{ij}(s)v_{ij}(\eta) \quad \text{for } \eta \in \Delta_i.$$  \hspace{1cm} (12)

Thus, the behavior of the cumulative wavenumber increment in the absorption cross-section is assumed independent of wavenumber and space. This assumption is not full-spectrum correlation, but rather, local-spectrum correlation within a spectral interval $\Delta_i$. The assumption represented by Eq. (12) is more mathematical than physical. It has the sense of simple separation of spatial and spectral variables in the function, which yields a difference of cumulative wavenumber at two adjacent cross-sections as $w(C_j, s, \eta) - w(C_{j-1}, s, \eta)$. This assumption is based on the observation that the cumulative wavenumber is not a rapidly growing function (at least not more rapidly growing than a linear function). The curves representing the cumulative wavenumber at two adjacent cross-sections are very similar, as seen in Fig. 2. Therefore, the difference in cumulative wavenumber is practically a constant with respect to $\eta$ if the spectral interval considered, $\Delta_i$, is not too large. Thus, the given assumption is an approximation. The smaller is the spectral interval $\Delta_i$ and the closer are the adjacent absorption-cross-sections, the better is the approximation. This local-spectrum correlation is much less restrictive than the widely used full-spectrum correlation techniques (e.g., ideal behavior of the spectrum in SLW modeling, fictitious gases ADF, full-spectrum correlated-$k$ distribution).
To define the function \( v_{ij}(\eta) \), choose some reference point \( s^* \) (at which we have reference values of temperature \( T(s^*) \) and species concentrations \( Y_m(s^*) \), including particles). One can arbitrarily assign for this location a value \( u_{ij}(s^*) \equiv 1 \). Then
\[
v_{ij}(\eta) = w(C_j, s^*, \eta) - w(C_{j-1}, s^*, \eta). \quad (13)
\]
For an arbitrary point \( s \), the function \( u_{ij}(s) \) can then be calculated by
\[
u_{ij}(s) = \frac{w(C_j, s, \eta) - w(C_{j-1}, s, \eta)}{w(C_j, s^*, \eta) - w(C_{j-1}, s^*, \eta)}. \quad (14)
\]
For the mixture of \( m \) gases of molar density \( N \) with particles (which have step-wise variation of absorption coefficient \( \kappa_{p,i} \) in the intervals \( \Delta_i \)), the function \( u_{ij}(s) \) according to Eq. (8) is defined by the following formula:
\[
u_{ij}(s) = \frac{\sum_m \left[ w_m(C_j/Y_m - \kappa_{p,i}/Y_mN, s, \eta) - w_m(C_{j-1}/Y_m - \kappa_{p,i}/Y_mN, s, \eta) \right]}{\sum_m \left[ w_m(C_j/Y_m - \kappa_{p,i}/Y_mN, s^*, \eta) - w_m(C_{j-1}/Y_m - \kappa_{p,i}/Y_mN, s^*, \eta) \right]}. \quad (15)
\]
The integration of the spectral intensity of radiation \( I_\eta \) over the fractional gray gas \( D_{ij} \) using the concept of cumulative wavenumber described by Eq. (3) yields:
\[
\int_{D_{ij}} I_\eta \, d\eta = \int_{\{\eta; C_{j-1} < C_j < C_j\}} I_\eta \, d\eta \\
= \int_{\{\eta; C_j < C_{j-1}\} \cap \Delta_i} I_\eta \, d\eta - \int_{\{\eta; C_j < C_{j-1}\} \cap \Delta_i} I_\eta \, d\eta \\
= \int_{\Delta_i} I_\eta \, dw(C_j, \eta) - \int_{\Delta_i} I_\eta \, dw(C_{j-1}, \eta) \\
= \int_{\Delta_i} I_\eta \, dw(C_j, s, \eta) - w(C_{j-1}, s, \eta) \\
= \int_{\Delta_i} I_\eta \, dw(u_{ij}(s)v_{ij}(\eta)) \\
= u_{ij}(s) \int_{\Delta_i} I_\eta \, dw(v_{ij}(\eta)) \\
= u_{ij}(s)J_{ij}(s), \quad (16)
\]
where \( J_{ij} \) is now viewed as a fractional gray gas intensity, and \( u_{ij}(s) \) can be viewed as a local correction factor to the fractional gray gas intensity. The total intensity of radiation is determined by the summation of all gray gas intensities weighted by the correction factor \( u_{ij}(s) \):
\[
I(s) = \int_0^\infty I_\eta(s) \, d\eta = \sum_{i,j} u_{ij}(s)J_{ij}(s), \quad (17)
\]
where the correction factor \( u_{ij}(s) \) is defined by Eqs. (14) or (15).
5. Cumulative wavenumber form of the RTE

The radiation field in absorbing and emitting media along a pathlength $s$ in a direction $\hat{\Omega}$ is described by the spectral intensity of radiation, the governing equation for which is the RTE [13]:

$$\frac{\partial I_\eta}{\partial s} = -\kappa_\eta I_\eta + \kappa_\eta I_{b\eta}.$$  

(18)

In a fashion similar to the derivation of Eq. (16), the spectral integration of Eq. (18) with the help of the cumulative wavenumber according to Eq. (3) leads to the set of equations for fractional gray gas intensities. The integration produces no Leibnitz terms because integration is performed not over $D_{ij}$ (which can be different for distinct spatial locations in non-uniform media) but over fixed spectral intervals $\Delta_i$:

$$\frac{\partial J_{ij}}{\partial s} = -\kappa_j J_{ij} + \kappa_j J_{bij},$$  

(19)

where $\kappa_j$ is the gray gas absorption coefficient, determined in this study from the supplemental absorption cross-sections as

$$\kappa_j = N \sqrt{C_j C_{j-1}}$$  

(20)

$J_{bij}$ is the fractional blackbody radiative energy source

$$J_{bij}(s) = \int_{\Delta_j} I_{b\eta}(T(s), \eta) \, dv_{ij}(\eta) = \int_{\Delta_j} I_{b\eta}(T(s), \eta) \, [w(C_j, s^*, \eta) - w(C_{j-1}, s^*, \eta)].$$  

(21)

The Planck radiation distribution function in Eqs. (19) and (21) is evaluated at the local temperature, and the function $v_{ij}(\eta)$ is evaluated at the thermodynamic state of the chosen reference location $s^*$ according to Eq. (13). The summation of $J_{bij}(s)$ over all fractional gray gases gives the total emission:

$$\sum_{i,j} J_{bij} = \sigma T^4/\pi.$$  

(22)

For non-gray, diffusely emitting and reflecting walls, the boundary condition is written as [13]

$$I_{\eta}(s_w, \hat{\Omega}) = \varepsilon_{\eta w} I_{b\eta}(T_w) + \frac{\rho_{\eta w}}{\pi} \int_{n \cdot \hat{\Omega}' < 0} I_{\eta}(s_w, \hat{\Omega}') |n \cdot \hat{\Omega}'| \, d\hat{\Omega}'$$  

(23)

where $\hat{\Omega}$ is the direction vector along the path $s$, $s_w$ defines the point on the boundary surface, and the index $w$ refers to quantities evaluated at the boundary; $\varepsilon_{\eta w}$ is the spectral emissivity, and $\rho_{\eta w}$ is the spectral reflectivity of the boundary. If we assume the step-wise variation in boundary optical properties over the spectrum, $\varepsilon_{\eta w} = \varepsilon_{iw}$ and $\rho_{\eta w} = \rho_{iw}$ for $\eta \in \Delta_i$, and integrate Eq. (23) over fractional gray gases $D_{ij}$, we receive boundary conditions for fractional gray gases intensities:

$$J_{ij}(s_w, \hat{\Omega}) = \varepsilon_{iw} I_{ijb}(T_w) + \frac{\rho_{iw}}{\pi} \int_{n \cdot \hat{\Omega}' < 0} I_{\eta}(s_w, \hat{\Omega}') |n \cdot \hat{\Omega}'| \, d\hat{\Omega}'$$  

(24)

When the fractional gray gas form of the RTE, Eq. (19), with boundary conditions, Eq. (24), is solved for all fractional gray gases $J_{ij}$, the total radiation intensity can be found by Eq. (17). In isothermal, homogeneous media, the correction factor $u_{ij}(s) \equiv 1$. 
6. Validation

6.1. Isothermal, homogeneous gas mixture with soot particles

Consider a mixture of soot with three combustion gas species, water vapor, carbon dioxide, and carbon monoxide. A plane-parallel slab of thickness $L = 1.0 \, \text{m}$ is filled with an isothermal homogeneous mixture of these three species at 1000 K and 1 atm total pressure. The mole fractions of H$_2$O, CO$_2$, and CO in the mixture are, 0.2, 0.1, and 0.03, respectively. The soot volume fraction in the mixture is $f_v = 10^{-7}$. The spectral absorption coefficient of the soot was approximated as a linear function of wavenumber according to $\kappa_{\text{soot}} = c f_v$, where $c$ was taken here to be 7.0 [2]. The boundaries are black and non-emitting. The fractional gray gas RTE, Eq. (19), was solved by the cumulative wavenumber (CW) method described above with 20 gray gases and a uniform spectral interval $\Delta \lambda = 100 \, \text{cm}^{-1}$ using the discrete ordinates method with 499 spatial grid points and 30 ordinates [6]. Fig. 5 shows the comparison of the CW model prediction of the local radiative dissipation source $Q(x)$ with line-by-line prediction (LBL) and SLW results [12] obtained previously employing the same spectral database, HITEMP [11]. The comparison shows very good agreement between the cumulative wavenumber method and the LBL benchmark solution. The results show that the CW method appears to be, in general, more accurate for this case than the SLW model.

6.2. Non-isothermal, homogeneous layer of CO$_2$

Consider a plane-parallel slab of thickness $L = 1.0 \, \text{m}$ filled with 10% carbon dioxide with uniform concentration and parabolic variation of temperature $T(x) = 1000 - 500(2x/L - 1)^2$. The walls are assumed to be black at 500 K. The fractional gray gases form of the RTE, Eq. (19), was solved with the discrete ordinates method employing 109 grid points and 12 ordinates. The radiative
dissipation source evaluated by the method outlined here based on the cumulative wavenumber and local-spectrum correlation (CW) using the HITEMP database with 10 gray gases is compared in Fig. 6 with LBL results obtained by Modest and Zhang [10]. The LBL results were carried out using the HITRAN96 spectral database. Although the two predictions use different spectral databases, the temperatures for this case ($T < 1000$ K) lie in a range where the absorption spectra in the HITEMP and HITRAN96 databases differ very little. The comparison between predictions shows good agreement.

Fig. 7 illustrates the sensitivity of predictions to the choice of reference location (and corresponding reference state). As shown, the choice of reference location $s^*$ anywhere inside the layer
Fig. 8. Predicted local radiative flux in non-isothermal layer of CO$_2$.

(including the boundaries) in the CW method gives no distinguishable difference in predicted profiles of the local radiative source, $Q(x)$. The maximum difference in predicted local flux divergence $Q(x)$ for the two extreme reference states employed ($T^* = 500$ and 1000 K) is under 2%. The local-spectrum correlation methodology developed here appears to offer considerable advantage over previous total-spectrum approaches in the minimization of sensitivity to the specification of reference state.

6.3. **Step-variation in temperature in a layer of CO$_2$**

Consider a plane-parallel slab of thickness $L = 1.0$ m filled with carbon dioxide with spatially uniform concentration of 30% (remainder nitrogen), and step-wise variation of temperature $T(x) = 1000$ K for $0 \leq x \leq 0.35$ m and $0.65 \leq x \leq 1.0$ m, and $T(x) = 1500$ K for $0.35 < x < 0.65$ m. Both walls were assumed gray and diffusely reflective with emissivity $\varepsilon = 0.8$ at 1000 K. The LBL and CW solutions were obtained with the same spectral database HITEMP [11], solving the RTE using the discrete ordinates method with 100 grid points and 16 ordinates. The CW simulations employed 20 gray gases. The LBL predictions were made by subdividing the interval between adjacent lines in the spectrum into eight subline intervals. The integration of the RTE assumed constant absorption cross-section and Planck blackbody intensity at the boundaries of each of these subline spectral intervals, and the trapezoidal rule was used for numerical integration. Further refinement yielded no appreciable change in LBL predictions. The predicted total local radiative flux is shown in Fig. 8. The agreement between LBL and CW predictions is very good even for this case where a discontinuity in layer temperature is modeled.
7. Conclusions

A new approach is presented for the prediction of radiative transfer in non-uniform mixtures of high temperature gases. The RTE is integrated with the help of a new gas absorption distribution function termed the cumulative wavenumber. Tabulated for the individual gas species, the cumulative wavenumber can be evaluated by a simple procedure for gas mixtures. The method allows, in the context of the weighted-sum-of-gray gases approach, the replacement of the integration over the spectral subintervals produced by the intersection of the supplemental cross-sections with the high resolution line molecular spectrum by the integration over fixed wavenumber intervals. Such an integration does not produce Leibnitz terms in non-uniform media (as in SLW modeling). The method preserves the advantages of the global WSGG and SLW methods, and it can be used with arbitrary RTE solvers. The method is efficient (computational times do not exceed 5 s on an HP C360 workstation). Local-spectrum correlation, used for the accommodation of non-uniform media, appears to be more justified than full-spectrum ideal behavior of the molecular spectrum. The predictions are independent of reference state used, and the model is well-suited for gas mixtures with non-gray particulates and boundaries.

References

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