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CHEMICAL KINETICS AT MICRO-, MESO-, MACROSCALES

A semi-empirical model for thermal decomposition of carbonates and its application to astrobiology

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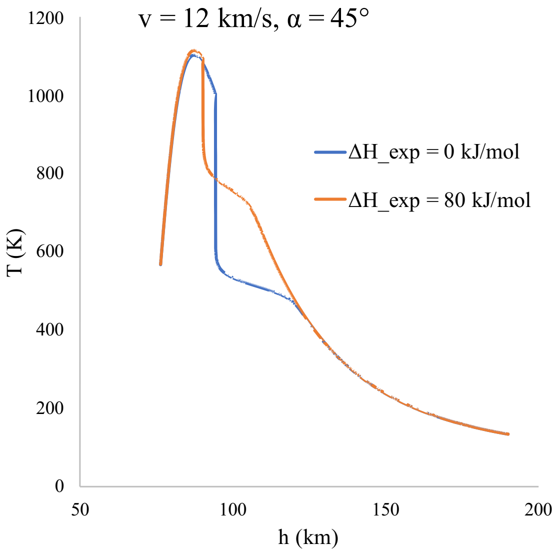
Abstract

We review the basis of a semi-empirical model of carbonate decomposition developed by the authors, shortly discussing numerous previous theories. Then, the model is applied to the modeling of a scenario of matter transport from space to the surface of planets embedded into sub-mm mineral grains, with the correction of the so-called additional enthalpy, coming from the experiments. Having magnesium and calcium carbonates as mineral phases and the atmosphere of Earth and Mars as environments, the chemical-physical history of grains entering at different angles and speeds is discussed. The results are compared with those obtained previously and new evaluations of the most promising scenarios are formulated.

Graphical abstract



Keywords Panspermia · Micrometeorids · Carbonates · Decomposition kinetics

1 Introduction

The discovery in the interstellar medium [i.e., Iglesias-Groth 2023; Rivilla et al. 2023] and in meteorites and other objects of the Solar System [i.e., Glavin et al. 1999; Hadraoui et al. 2019] of increasingly complex organic molecules leads to a growing interest in the study of hypothetical scenarios in

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Extended author information available on the last page of the article which organic molecules of space origin contribute to or influence the origin of life on our planet.

A weaker panspermia scenario, in which the external environment contributes with molecules such as amino acids or nucleotides, and a stronger one, involving the spatial transfer of life forms in some quiescent state, require the formulation of chemical-physical hypothesis for the transport of this material through the interplanetary medium. This medium is known to be aggressive for biological materials, even for simple organic molecules. One of the most considered hypotheses is the one of transport in a mineral phase in micrometeoroid, which can provide protection from radiation and survive the atmospheric entry. This hypothesis is supported by the presence of organic molecules in meteorites [Ehrenfreund and Charnley 2000; Botta and Bada 2002; Sephton 2005].

In this framework, theoretical models have been developed [Love and Brownlee 1991; Campbell-Brown and Koschny 2004; Briani et al. 2013; Micca Longo and Longo 2021] and ambitious experiments have been carried out in the context of international collaborations. For example, the STONE experiments explicitly addressed the ability of mineral samples to protect organic substances using a payload placed outside an atmospheric entry vehicle from low Earth orbit (LEO) [Brack et al. 2002; Parnell, et al. 2008; Foucher et al. 2010; Parnell et al. 2011]. Among the materials considered in this experiment, carbonates play a crucial role. On the Earth, carbonates are associated with life forms: this represents an important breakthrough compared to previous literature, that has always considered mineral phases typical of those of meteorites, i.e. magnesium and iron silicates [Froh, et al. 2023]. Furthermore, these minerals are present within the well-known Martian meteorite ALH84001, in the form of nodules with an extremely rich association of organic molecules [McKay et al. 1996; Friedmann et al. 2001; McKay et al. 2009; Thomas-Keprta, et al. 2009]. More recently, *in-situ* analyses of rock samples, taken from the floor of the Martian Jazero crater by Perseverance rover, have shown the presence of carbonate-bearing minerals in association with aromatic organic compounds, most likely single- and double-ring aromatics [Scheller et al. 2022].

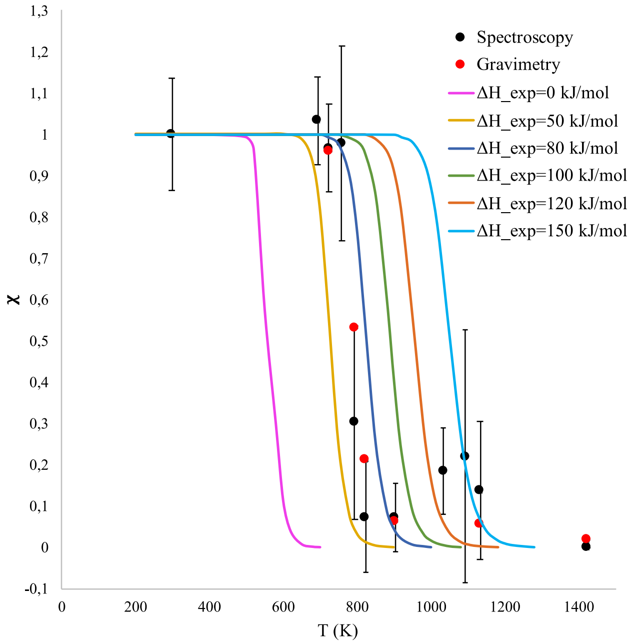

Fig. 1 Comparison between the theoretical values (solid lines) of the χ parameter and the experimental ones (circle dots) obtained both by gravimetric and spectroscopic techniques, for a $CaCO_3$ micrograin. Figure adapted from [Micca Longo et al. 2019]

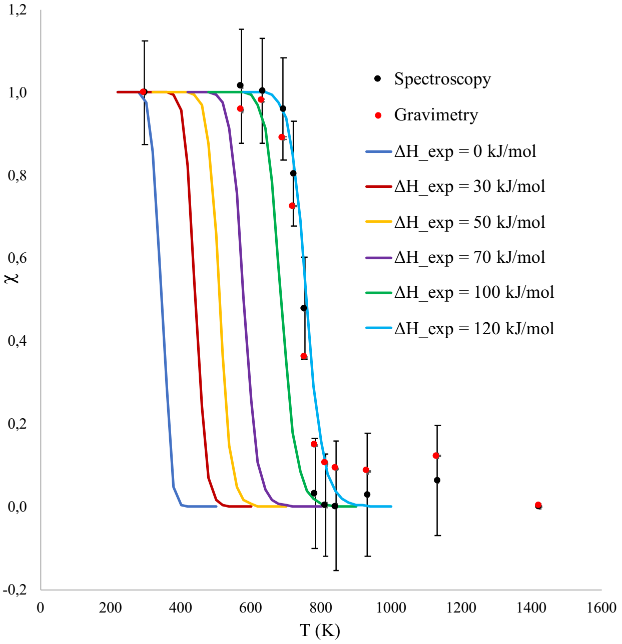

Fig. 2 Comparison between the theoretical values (solid lines) of the χ parameter and the experimental ones (circle dots) obtained both by gravimetric and spectroscopic techniques, for a $MgCO_3$ micrograin. Figure adapted from [Mancarella et al. 2022]

The development of computer simulation models has been underway for many years to study scenarios of thermal protection produced by grains of bio-associated minerals, such as carbonates and sulfates [Micca Longo and Longo 2021], and to overcome the limitations of experiments that cannot reproduce the authentic entry speeds: an object in LEO moves at 7.8 km/s, while one reaching the Earth's atmosphere from the interplanetary space cannot be slower than 11.2 km/s, the Earth's escape velocity. These two quantities are sometimes addressed as first and second cosmic velocities, respectively [Arnol'd 2013]. Furthermore, computer models make it possible to extend the study to the ancient Earth's atmosphere at the time of the origin of life [Micca Longo and Longo 2020] and to other planets such

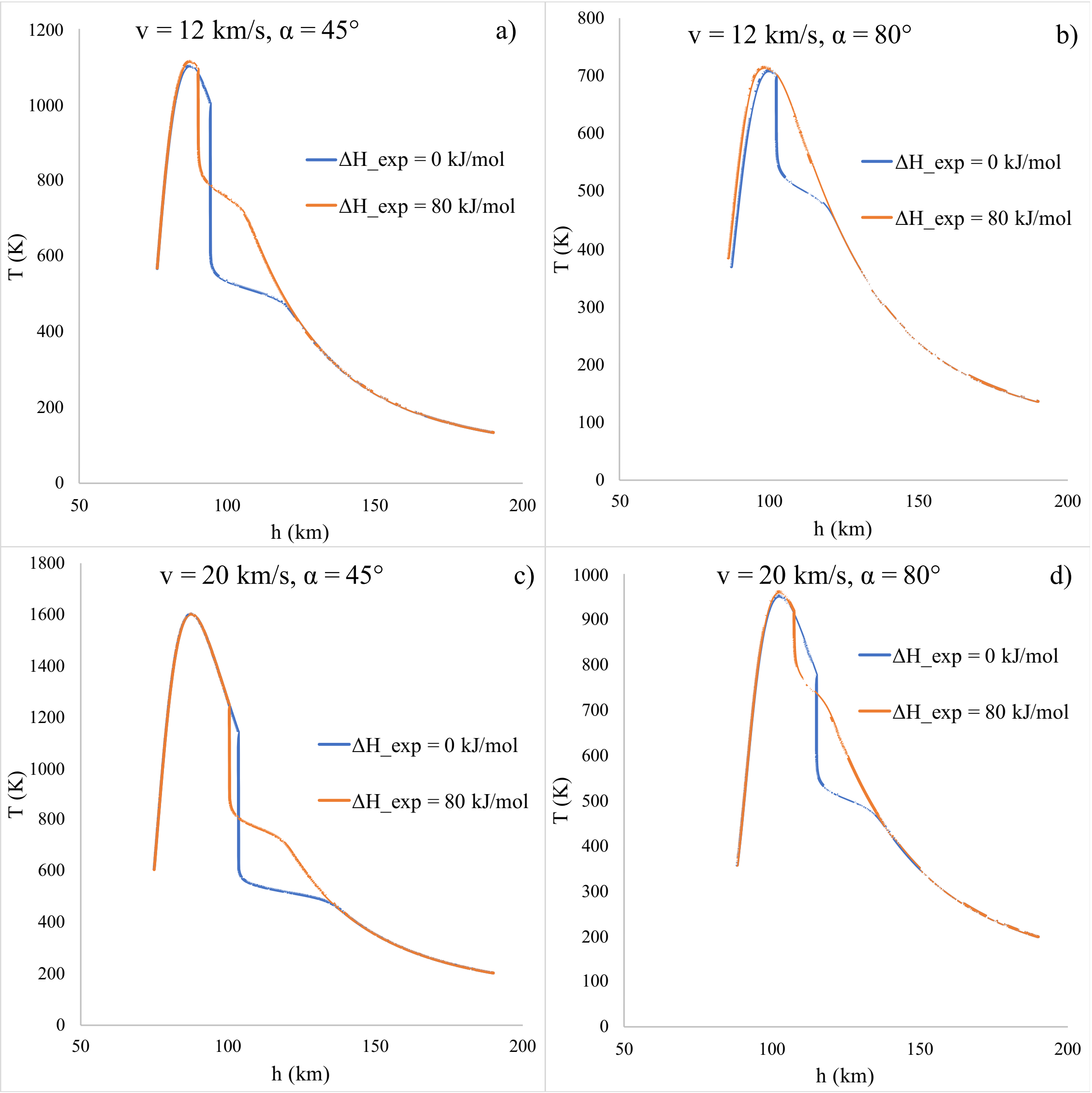

Fig. 3 Thermal history of a $CaCO_3$ micrograin entering the Earth atmosphere

as Mars [Micca Longo and Longo 2022]. These simulations highlight an aspect not considered in previous studies: the role played by the thermal decomposition reaction of these minerals:

$$(Ca, Mg)CO_3 \rightarrow (Ca, Mg)O + CO_2$$

This process proceeds rapidly even at moderate temperatures, for atmospheric entry standards; furthermore, it is endothermal, so it leads to temperature reduction during the passage through the atmosphere [Micca Longo and Longo 2021].

More recent works have validated the kinetic-chemical part of these input models, using decomposition experiments of carbonate grains of a size compatible with the scenario and performed in a vacuum furnace [Micca Longo et al. 2019; Mancarella et al. 2022]. The comparison with experiments showed that the theoretical model based on Langmuir's law is qualitatively valid, but it can be improved with the addition of a further Arrhenius-type factor, with the threshold energy determined by the comparison between the chemical kinetic model and experiments.

In this work for the first time, we present the results of a model of entry into the atmosphere of grains of two carbonates, which are promising from the prebiotic point of view: this model employs a formulation of the chemical kinetics of thermal decomposition improved with the addition of the experimentally determined term. The results provide new insights into the most promising scenarios for both Earth and Mars.

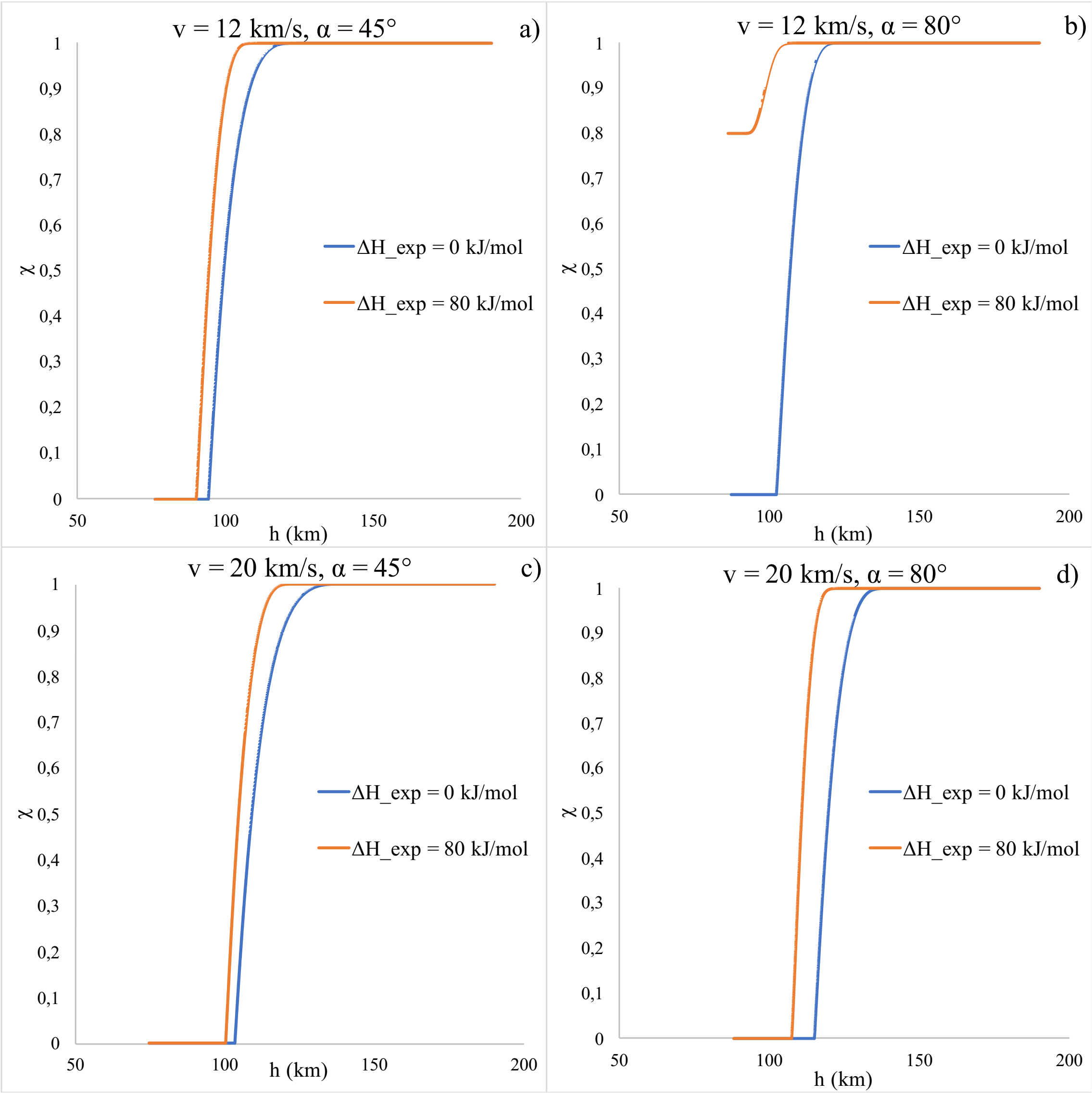

Fig. 4 Carbonate fraction of a $CaCO_3$ micrograin entering the Earth atmosphere

2 Kinetic model of the thermal decomposition

The key point of our model lies in the equation for the loss rate J (mole m^{-2} s^{-1}) of the fraction of the volatile component of the mineral phase, that is the carbonate fraction χ. It can be written as:

$$J = -\chi \frac{1}{4}\sqrt{\frac{8RT}{\pi M}}\frac{p}{RT} \tag{1}$$

This is essentially Langmuir's law for evaporation [Langmuir 1916], based on the chemical kinetics of gases and on the principle of detailed balance, corrected by Raoult's law [Bisceglia et al. 2017] to account for the changing composition of the mineral in the course of the decomposition process. The first term, the mole carbonate fraction χ, appears in the equation in the hypothesis that the carbonate-oxide mixture is an ideal one. The remaining factors give the molar rate of gas molecules hitting the solid unit surface per unit time at equilibrium, resulting from elementary kinetic theory [Loeb 2004]: it includes the geometric factor ¼, the expression of the mean thermal speed of the gas component and the number of moles of gas per unit volume, again at equilibrium. R is the ideal gas constant, T is the temperature, M is the molar mass of the gas component (CO_2, in this case). p is the vapor pressure of a pure carbonate at a given temperature T given by:

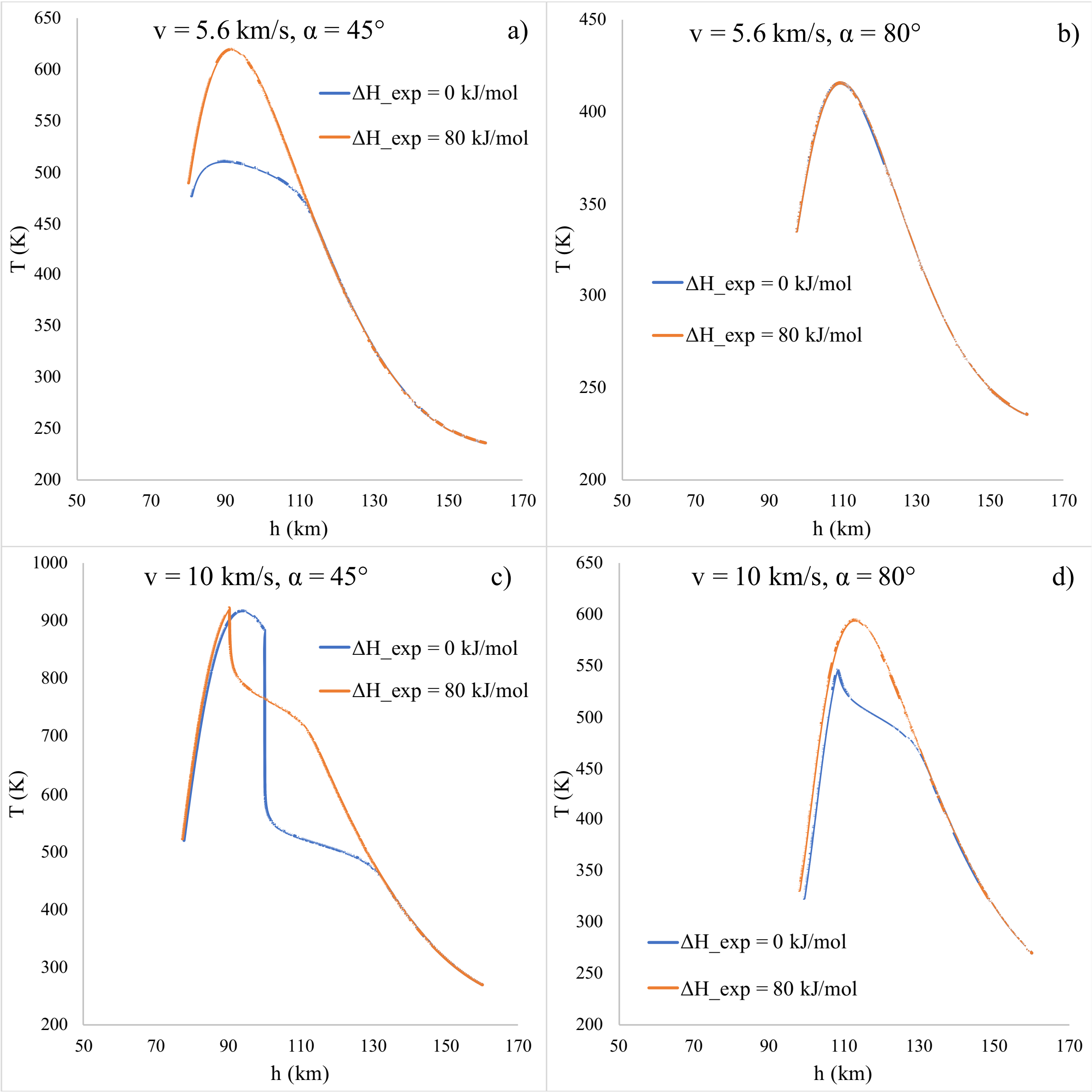

Fig. 5 Thermal history of a $CaCO_3$ micrograin entering Mars atmosphere

$$p = p_0 e^{-\frac{\Delta G_0(T)+\Delta H_{exp}}{RT}} \tag{2}$$

where p_0 is the reference pressure for the thermodynamic data, $p_0 = 1.01 \times 10^5$ Pa. $\Delta G_0(T)$, the Gibbs free energy of the reaction, is calculated using the specific thermodynamic data of enthalpy ΔH_f and entropy ΔS_0 of the chemical species involved in the decomposition process. The innovation of the model is the term ΔH_{exp}, that represents an additional kinetic term that can introduce a sort of energy threshold in the evaporation process: we call it *additional enthalpy*. The reason for this denomination is that in this way Langmuir's law remains formally valid. Being a threshold, it is not considered in the thermal energy balance. This quantity is determined by a comparison between the theoretical model and the experiment.

These experiments [Micca Longo et al. 2019; Mancarella et al. 2022] were carried out by heat treatment in a vacuum furnace of a carbonate particulate sample (calcium carbonate or magnesium carbonate) with a known grain size. After the heating process, the mole fraction of carbonate was determined by gravimetry and by infrared spectroscopy. As shown in Figs. 1 and 2, the decomposition law provided by the model agrees with the experiments if the additional enthalpy ΔH_{exp} is introduced: the ΔH_{exp} values for $MgCO_3$ grains and for $CaCO_3$ ones are 120 kJ/mol and 80 kJ/mol respectively, both for grains with an estimated size range of $25-50\ \mu$m. This additional enthalpy might be related to the presence of barriers to the CO_2 diffusion, or to additional

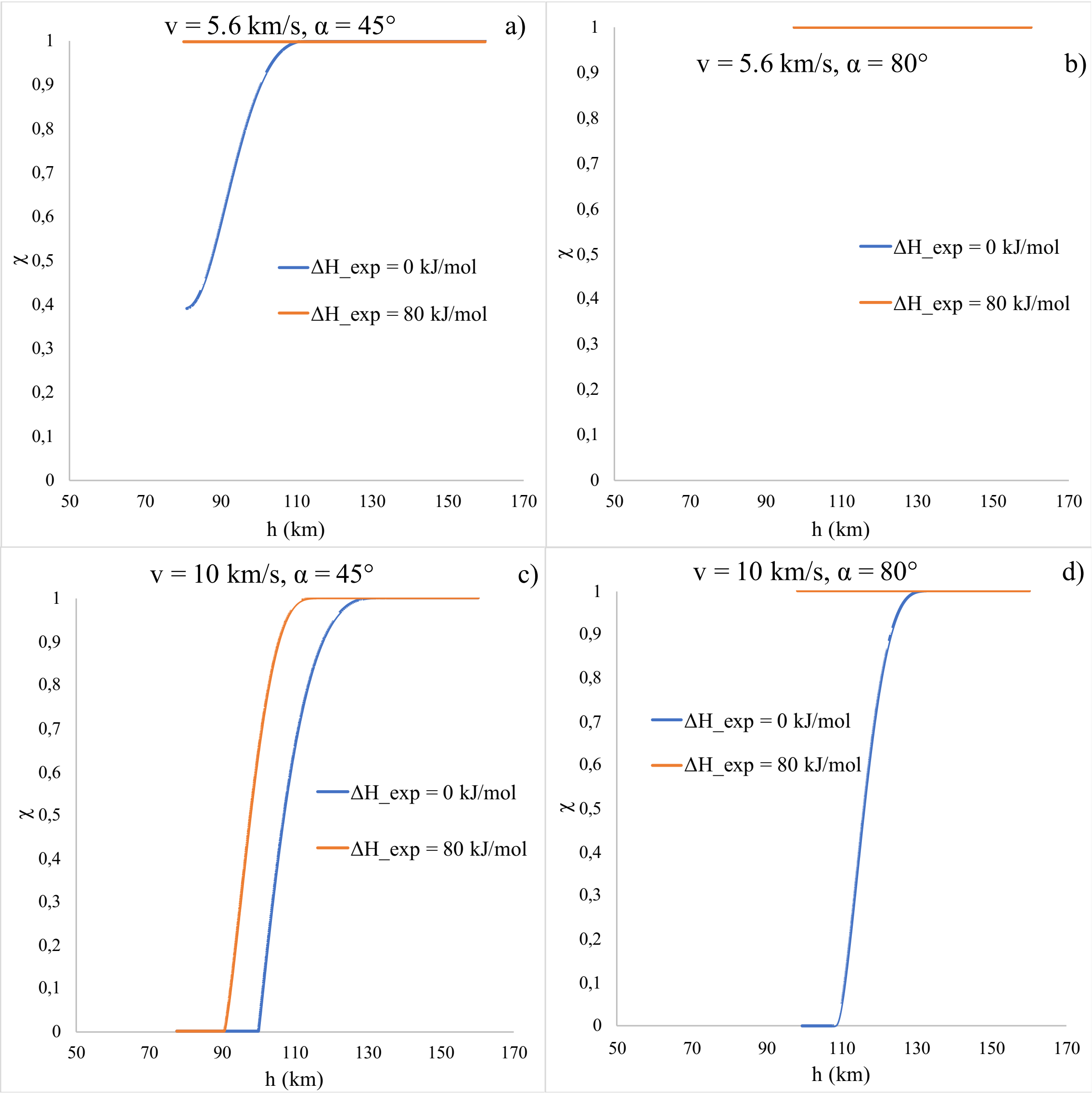

Fig. 6 Carbonate fraction of a $CaCO_3$ micrograin entering Mars atmosphere

surface energy associated with crystal breaking when CO_2 is released.

It should be noted that Langmuir's law excellently reproduces the rate of evaporation of pure solid materials and those that evaporate completely into gaseous components: the difficulty in the case of carbonates arises from the fact that the process leaves behind a solid fraction, the oxide, which is not removed. This kinetic hindrance has been discussed in previous works and explained as an effect of the porosity of the material [Barker et al. 1973; Beruto, et al. 1980] or the formation of an onion structure, with oxide on the outside [Vosteen 1971]. Since this hindrance mechanism is certainly influenced by the grain size, the additional enthalpy determined by comparison with experiments is valid only for that precise grain size. Consequently, we use the semi-empirical method only for scenarios of entry into the atmosphere of grains that have the same size as that used in the laboratory.

3 Entry model and previous results

The differential equation described in the previous section must be inserted into a model that describes the entry into a planet's atmosphere of small grains of the materials mentioned, to determine promising scenarios for the dissemination of organic material.

The atmospheric entry model is a multi-physical one since it includes aspects of dynamics, thermodynamics, atomic mechanics, and heat transport. The model includes

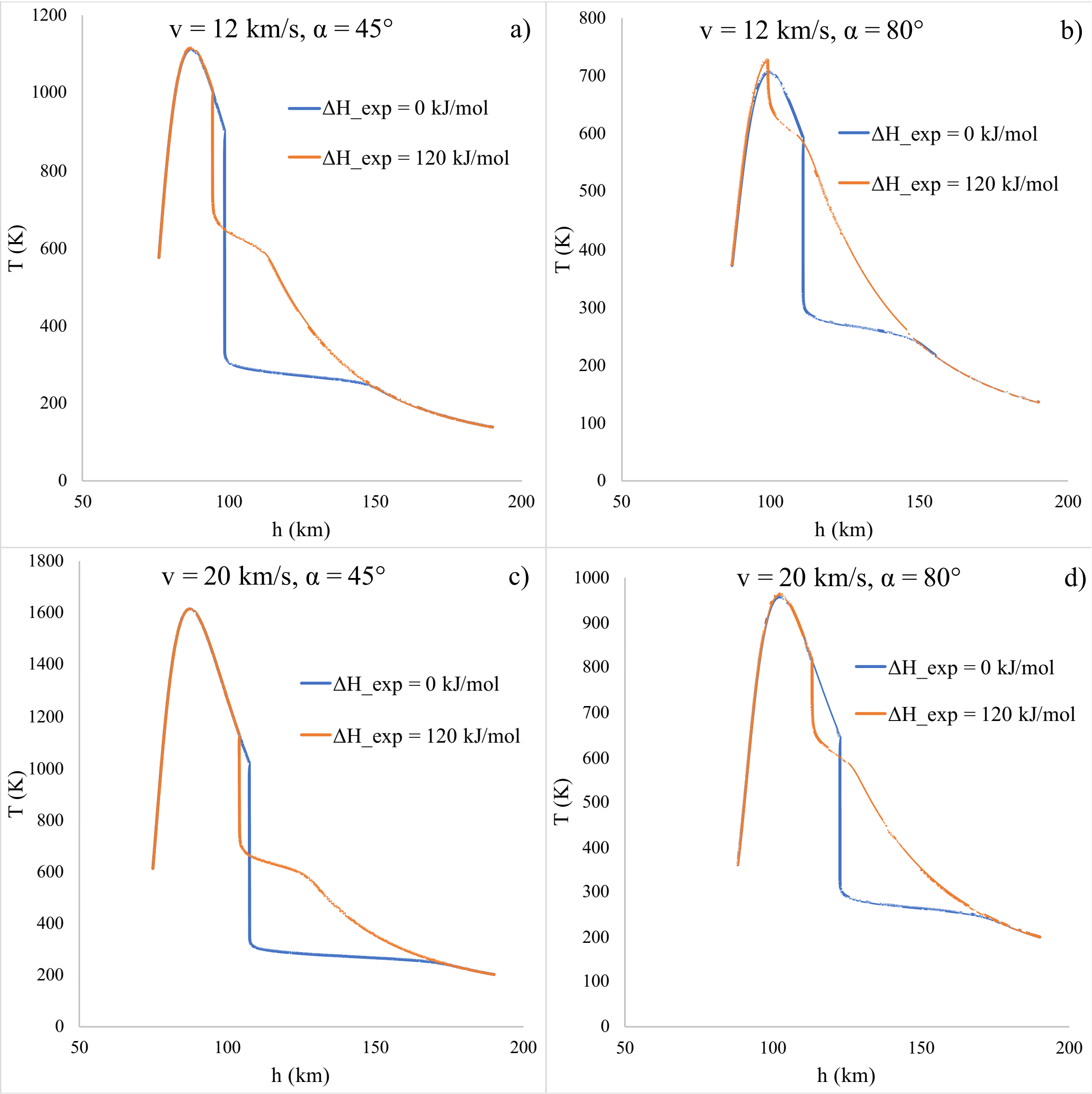

Fig. 7 Thermal history of a $MgCO_3$ micrograin entering the Earth atmosphere

numerous variables, such as the mass of the grain, its entry speed and entry angle, temperature and chemical composition [Micca Longo and Longo 2021]. The variations in time of these quantities are described in terms of differential equations of physical laws. For example, the velocity of the grain is influenced by gravity and by the momentum transmitted by the molecules of the atmosphere that impact the grain, the so-called atmospheric friction. The temperature *T* of the grain is described by the thermal energy flows and essentially depends on three aspects: the transfer of kinetic energy by the atmospheric molecules, the cooling due to black body radiation, and the cooling due to the endothermal decomposition. The detailed model has been described in previous publications [Micca Longo and Longo 2021; Micca Longo and Longo 2022] and is basically the well-known Love and Brownlee model [Love and Brownlee 1991].

The behavior observed in the simulation basically consists of a sudden slowdown of the grain when the atmospheric density becomes sufficiently high. Significant warming is observed at this slowdown. For some minerals, depending on the entry conditions, the mineral phase can decompose into carbon dioxide and metal oxide, which is refractory and does not melt even at the highest temperatures reached in these scenarios. This process absorbs energy and contributes to the temperature decreasing. When the decomposition is total, the scenario is not significant from the point of view of the potential transport of organic matter from space. The most interesting scenarios are observed for low entry speeds and for entry angles as close to the horizontal as possible (not

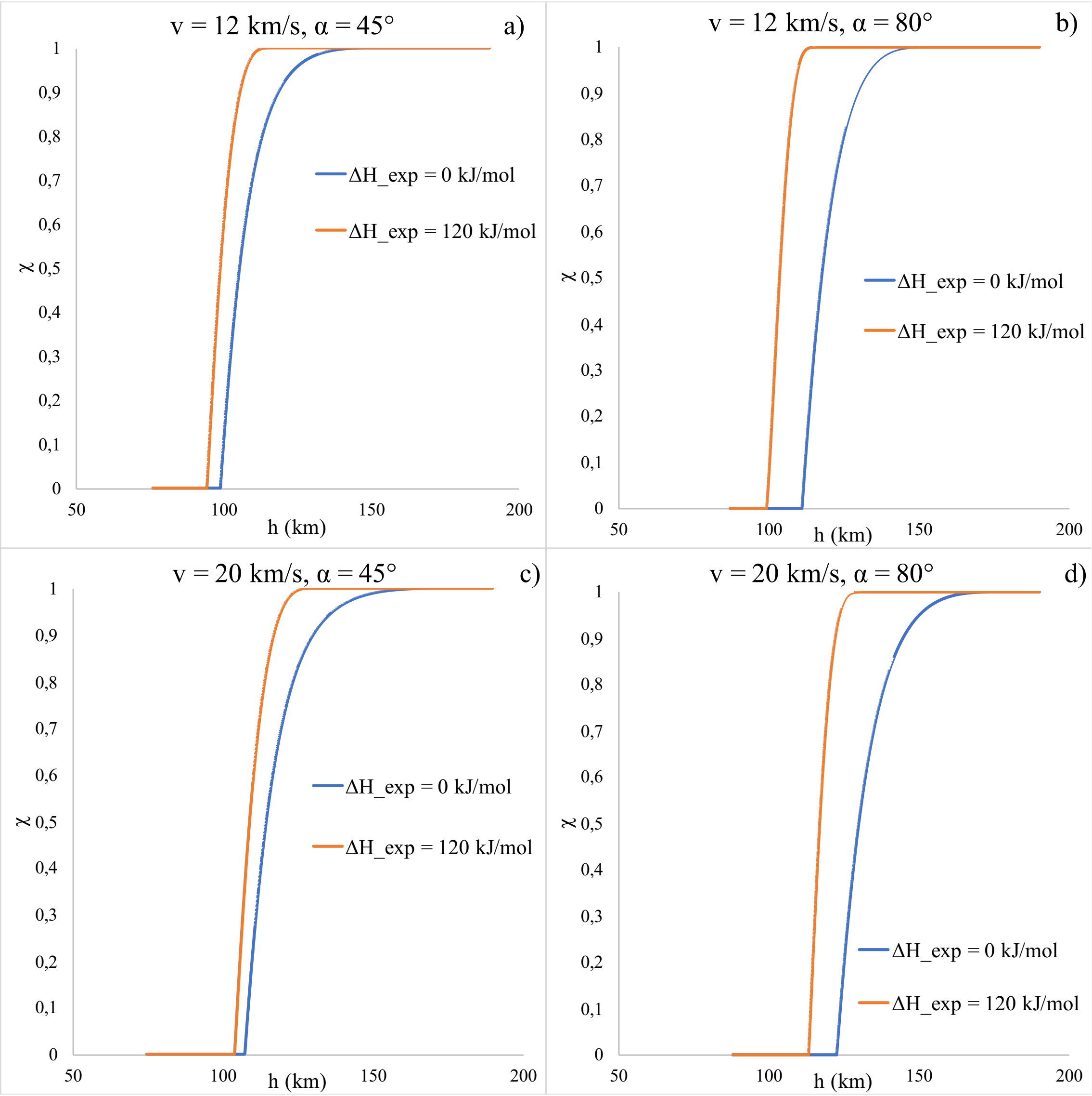

Fig. 8 Carbonate fraction of a $MgCO_3$ micrograin entering the Earth's atmosphere

so close as to allow the grain to bounce off the atmosphere and escape back into space). Since the minimum speed that a grain can have at the entrance in the atmosphere corresponds to the escape velocity from the planet, a mild entry is easier to obtain in the case of the Martian atmosphere than in the case of the Earth's atmosphere. Indeed, the lower escape velocity has suggested the hypothesis that Mars may have been a collector of organic matter of interplanetary origin in the past [Tomkins et al. 2019; Wilson et al. 2019; Micca Longo and Longo 2022].

In our previous works, the kinetic law for χ did not include the empirical quantity ΔH_{exp} described above. In the next section, we discuss the impact of this experimentally determined factor during the micrometeoroid atmospheric entry.

4 Results and discussion

Several entry scenarios concerning calcium carbonate and magnesium carbonate micrometeoroids, with a diameter of 50 μm, are presented. Thermal histories regarding micrograins' atmospheric entry allow to focus on their thermal behavior, peak temperature, and chemical decomposition.

In Figs. 3, 4, 5, 6, 7, 8, 9, 10, we show the thermal and chemical evolution (in terms of carbonate fraction χ) of $CaCO_3$ and $MgCO_3$ micrograins entering the terrestrial or Martian atmosphere with different velocities and entry angles. In these figures, that highlight the effect of the empirical additional enthalpy ΔH_{exp} on both thermal and chemical histories of such grains, the blue curve corresponds to the model of our previous works, while the orange one is

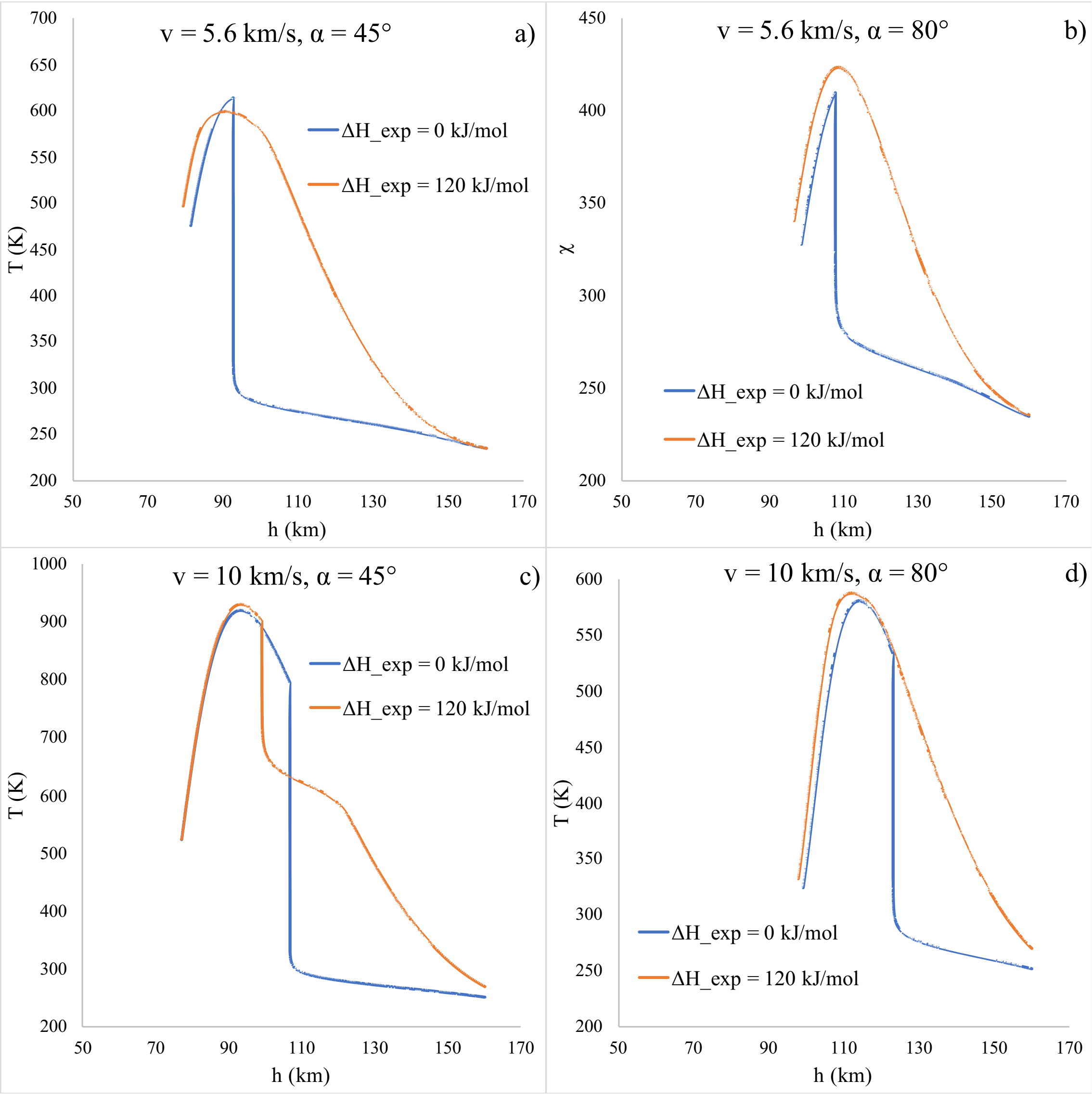

Fig. 9 Thermal history of a $MgCO_3$ micrograin entering Mars atmosphere

obtained with Eq. 1 for the mineral decomposition kinetics. The differences between the previous and the new results concern both the thermal and the chemical history of the grain. These differences can be understood by taking into account the effect of the new factor on the decomposition kinetics.

The temperature curve of a carbonate grain has a complex trend, characterized by various stages, starting from the initial phases of the process. As can be seen for example in Fig. 3, the initial increase in temperature quickly leads, if the temperature rises sufficiently, into the *plateau stage* in which the temperature remains almost constant: this behavior is a peculiarity of volatile minerals and depends on the fact that the temperature is mostly controlled by the phase transition between carbonate and oxide. If the carbonate is exhausted, that is the typical outcome for entry into the Earth's atmosphere, a rapid rise in temperature is observed, followed by the thermal peak in the phase of greatest deceleration of the grain.

For example, Fig. 3a refers to the entry into the Earth's atmosphere of a grain of $CaCO_3$ at a speed slightly higher than the escape speed and at an intermediate entry angle: the new model leads to a higher temperature of about 200 K in the plateau stage (~ 100 km), that is controlled by mineral decomposition. This is what is expected by observing that the onset of the decomposition in the experiments with $CaCO_3$ is about 200 K higher than the prediction of the Langmuir model (Fig. 1). The higher plateau temperature is however compensated by a shorter thermal peak, as can be seen from Figs. 3a, c, d where the onset of

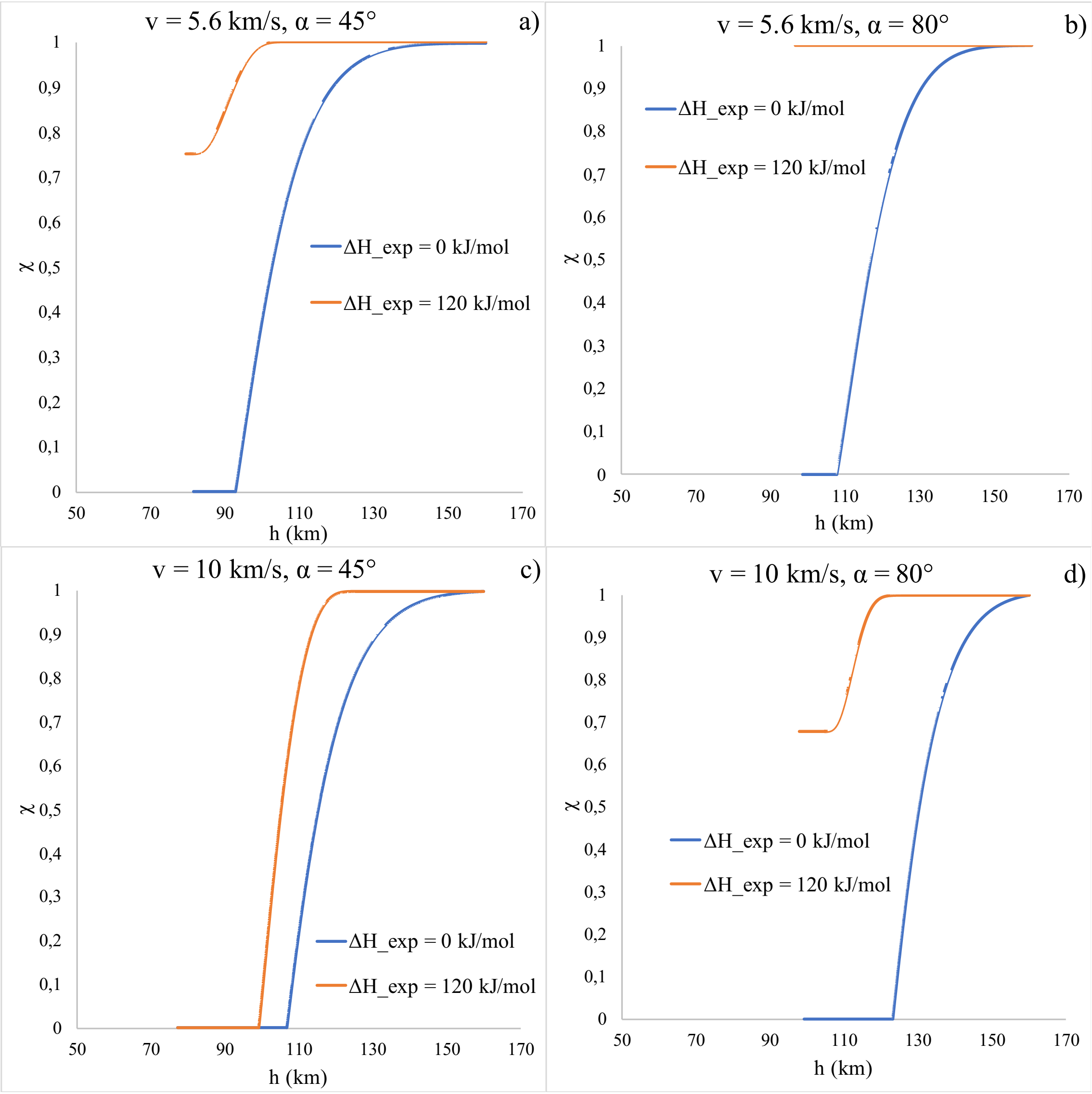

Fig. 10 Carbonate fraction of a $MgCO_3$ micrograin entering Mars atmosphere

the peak is shifted at lower altitudes and therefore its duration is reduced. This effect is relevant since the thermal peak is critical for the survival of organic matter associated with the grain. In a different case, like in Fig. 3b, the delay in the decomposition only increments the temperature throughout, which however is rather low during the whole trajectory. Observing Fig. 5a, which refers to a grain of $CaCO_3$ entering the atmosphere of Mars, once again at a speed slightly higher than the escape velocity, we see that the new kinetic model simply predicts an increase in the temperature of a peak still around 200 K. Even if this scenario is quite promising from a thermal point of view, it is not significantly compromised as the temperature still remains moderate at around 600 K. Regarding the chemical history of the mineral, we observe in all cases a slowing down of the decomposition kinetics and a consequent greater fraction of volatile components at a given altitude.

The difference is critical in a particular group of results: for example, in Fig. 6, the new results lead to an almost total survival of the volatile part of the mineral down to the ground when the old results produced a total decomposition. Obviously, in cases where the decomposition of the mineral was not significant in the previous results, such as Fig. 5b already mentioned, the new decomposition model does not produce visible differences.

A particularly interesting case is plotted in Fig. 7b, where the new scenario of a $MgCO_3$ grain in the Earth's atmosphere is able to significantly reduce the duration of the temperature peak. This is a “grazing” entry scenario at an almost horizontal angle, notoriously the most promising from an astrobiological point of view [Briani et al. 2013; Micca Longo and Longo 2021]. These aspects require the formulation of chemical kinetics of the decomposition of organic components: a rather complex problem that has so far only been addressed in a recent work [Canepa 2020].

The additional enthalpy introduced in the new decomposition model, as mentioned previously, is an additional factor that represents a barrier to chemical decomposition, whose nature has yet to be determined. The effect is likely to be due to the slow diffusion of carbon dioxide from the solid. SEM photographs of the material before and after the decomposition process, shown in our previous work [Micca Longo et al. 2019] and in [Powell and Searcy 1982], seem to suggest that part of this barrier is due to an energetic impediment associated with the structural transformation of the material during gas evolution. In this case, the additional enthalpy should also be added to the cooling terms in the grain temperature equation in the overall model: this would produce even more favorable entry scenarios.

5 Conclusions

In this work, for the first time we have applied Langmuir-Raoult's thermal decomposition kinetics with an empirical Arrhenius correction factor to a model of atmospheric entry of microparticles composed of calcium and magnesium carbonates. The additional enthalpy is set on the basis of experimental results, for grains of the same composition and size as those considered in the model. In this way, we have for the first time removed a limitation of the input model repeatedly reported in our previous works, and we now have a decomposition kinetics as reliable as is possible in light of current knowledge. The additional enthalpy introduced into the model produces several differences compared to previous results, in both the grain's thermal history and its chemical history. In some specific cases, these effects and differences deserve to be discussed with regard to defining a scenario of deposition of organic matter of interplanetary origin, on Earth and Mars. However, the results of the model, in their overall suggestion of an interesting scenario of organic matter protection, are not altered in a qualitative way by the new decomposition model. This implies that while the new model is fully compatible with the experiments and takes into account kinetic barriers to decomposition, the numerous results we have published in the past regarding the astrobiological interest of these scenarios are substantially confirmed.

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Data availability The model and thermodinamic data are available by contacting the authors.

Declarations

Conflict of interest The authors declare no competing interests.

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