Numerical simulation of rapid expansion of supercritical fluid

H. KSIBI *1, C. TENAUD **, P. SUBRA and Y. GARRABOS ***

ABSTRACT. – The expansion of a supercritical fluid solution through a nozzle is the key to one approach for producing microscale particles. It is based on the hyper-compressibility and the change of the solution capacity of the fluid from supercritical fluid conditions (high pressure) in expansion to a lower pressure environment. Simulation of the flow needs a good approximation of the Riemann problem to this type of fluid, and an accurate representation of the supercritical state by a suitable Equation of State (EOS), the integration of the equations of the motion is performed with the implicit TVD scheme of Harten and Yee and solved with the Alternating Direction (ADI) method.

Comparison with several test problems is given. It shows the reliability of the scheme for capturing shock waves and rarefaction waves, for representing the gradients of different variables in the nozzle and the jet, compression and expansion phenomena in the jet, and behaviour of the flow near the critical region.

1. Introduction

At the present time, research related to the use of supercritical fluids is spreading rapidly in many countries. Among these uses, the Rapid Expansion of Supercritical Solution (RESS), because of its potentialities, is being widely studied in the pharmaceutical, ceramic, food processing and other industries.

Supercritical conditions refers to the state of a substance when its critical point (critical temperature and pressure) has been exceeded (Fig. 1). Fluids in such a condition have a liquid-like density and a viscosity close to the viscosity of gas. They exhibit high solution capability towards many substances, combined with high rates of mass transfer.

The production of particles upon depressurization of supercritical fluid solutions was first noted over a century ago (Hannay et al., 1879). The formation of particles during the expansion of supercritical solutions through a nozzle has more recently been studied in many experiments (Tom and Debenedetti, 1991; Ksibi et al., 1995). We are currently investigating the RESS process, from an hydrodynamic point of view, in order to control the nucleation and particle growth during the expansion.

* LIMHP CNRS, Université Paris XIII, avenue J. B. Clément, 93430 Villetaneuse, France.
** LIMSI CNRS, Université de Paris XI, 91403 Orsay, France.
*** ICMCB CNRS, Université de Bordeaux I, Château de Brivazac, 33600 Pessac, France.
1 Present adress: 12, place 2 Mars, Sfax 3000, Tunisia.

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In this work, a numerical code has been developed to study the dynamics of the supercritical fluid expansion. This code solves the full, time dependent, Navier-Stokes and energy equations using a specific equation of state (EOS) proposed by Altunin et al. (1971). This set of equations is solved by using the Total Variation Diminishing (TVD) algorithm, introduced by Harten et al. (1986) and Yee et al. (1987), which is suitable for transonic and supersonic flows. To improve the spatial accuracy, conservative shock-capturing schemes must be of second order. Since we look for a steady solution, we want to use as large a time step as possible; in this way, the convergence is accelerated by implicit linearisation using the ADI formulation. These schemes are fairly efficient and accurate for two-dimensional supersonic and viscous flows.

To capture shock waves and discontinuities, we use an approximate Riemann solver developed by Vinokur and Montagné who adapted Roe’s averaging to a nonconvex EOS for a real gas (dense gas in our case). The determination of the invariants in the Riemann problem involves interpolation expressions for the thermodynamic variables: heat capacity at constant volume \( (C_v) \), thermal factor at constant volume \( (\alpha_v) \), and the derivatives of pressure, which are easily calculated, as will be seen hereafter.

In the following, we look at the conservation laws with the EOS from Altunin and Gadetskii. The coordinate transformation as well as the computational domain are then described. Jacobian matrices of the two Euler fluxes and their eigenvectors and eigenvalues are then given. We describe the approximate Riemann solver for the solution of these equations, the TVD algorithm and the boundary conditions. Finally we display and discuss numerical results obtained for this problem with three different test cases.
2. Numerical methods

2.1. Equations of the flow

The Navier-Stokes equations governing the unsteady, compressible, viscous fluid flow, in an axisymmetric domain may be written in conservative form as:

\[
\frac{\partial(yQ)}{\partial t} + \frac{\partial(yF)}{\partial x} + \frac{\partial(yG)}{\partial y} = 0.
\]

Here, \(x\) represents the longitudinal axis, \(y\) refers to the radial axis; \(Q\) is the solution vector; \(F\) and \(G\) can be decomposed as

\[
(2) \quad F = F_1 - F_v
\]

\[
(2') \quad G = G_1 - G_v.
\]

In (2, 2'), \(F_1\) and \(G_1\) corresponding to the Euler part of the terms \(F\), \(G\) while \(F_v\) and \(G_v\) refer to the viscous part:

\[
Q = \begin{pmatrix} \rho \\ \rho u \\ \rho v \\ \rho E \end{pmatrix}, \quad F_1 = \begin{pmatrix} \rho u \\ \rho u^2 + P \\ \rho u v \\ \rho u (\rho E + P) u \end{pmatrix}, \quad G_1 = \begin{pmatrix} \rho v \\ \rho v u w \\ \rho v^2 + P \\ \rho v (\rho E + P) v \end{pmatrix}
\]

\[
F_v = \begin{pmatrix} 0 \\ \tau_{xx} \\ \tau_{xy} \\ u \tau_{xx} + v \tau_{xy} + \kappa \frac{\partial T}{\partial x} \end{pmatrix}, \quad G_v = \begin{pmatrix} 0 \\ \tau_{xy} \\ \tau_{yy} \\ u \tau_{xy} + v \tau_{yy} + \kappa \frac{\partial T}{\partial y} \end{pmatrix}
\]

where

\[
(6) \quad \tau_{xx} = \frac{2}{3} \mu \left( 2 \frac{\partial u}{\partial x} - \frac{\partial v}{\partial y} \right), \quad \tau_{yy} = \frac{2}{3} \mu \left( 2 \frac{\partial v}{\partial y} - \frac{\partial u}{\partial x} \right), \quad \tau_{xy} = \tau_{yx} = \mu \left( \frac{\partial u}{\partial y} + \frac{\partial v}{\partial x} \right).
\]

Here \(\rho\) is the density, \(u\) and \(v\) are the velocity components in the \(x\) and \(y\) directions respectively, and \(E\) represents the total energy. The coefficients of viscosity and thermal conductivity \(\mu\) and \(\kappa\) respectively are written (Olchowy and Sengers, 1988) as follows (composed of two additive contributions due to temperature \(T\) and density \(\rho\)):

\[
(7) \quad \kappa = \kappa_0(T) + \kappa(\rho)
\]

\[
\mu = \mu_0(T) + \mu(\rho),
\]

where
\( \kappa_0(T) = \sqrt{T} [\kappa_0 + (\kappa_0/T)]^{-1} \)
\( \kappa(\rho) = \kappa_1 \rho + \kappa_2 \rho^2 + \kappa_3 \rho^3, \)

and

\( \mu_0(T) = \sqrt{T} \cdot 10^{-7} [\mu_0 + (\mu_0/T)]^{-1} \)
\( \mu(\rho) = \mu_1 \rho + \mu_2 \rho^2 + \mu_3 \rho^3. \)

The quantities \( \mu_0(T) \) and \( \kappa_0(T) \) are the dilute gas transport coefficient limits at the same temperature \( T \). The expressions for \( \mu(\rho), \kappa(\rho) \) are good approximations for the density dependency; \( \mu_{ij}, \kappa_{ij}, \mu_i \) and \( \kappa_i \) are constant coefficients.

2.1. Equations of State and Thermodynamic Factors

Most equations of state are valid only for part of the experimentally-studied region of change in parameters; Borel (1991) has discussed several equations of state, such as the equations proposed by Van Der Waals, Peng-Robinson and Redlich-Kwong. We reported in Figure 2 the evolution of pressure versus the density for \( \text{CO}_2 \) using both the previous analytical EOS (listed above) and the IUPAC’s experimental data (Angus et al., 1975) for a given temperature. We note that calculation and experimental values are in good agreement up to the critical region. However, in the supercritical region (high pressure, high density), important increases in pressure errors are noted when comparing calculation and experimental measurements. We observe the same behaviour at higher temperatures. Therefore, there is no chance of describing the behaviour correctly using these equations within the supercritical domain.

Fig. 2. – Comparison of different EOS for Carbon Dioxide at a given temperature (325 K). (PG: Perfect Gas EOS, VDW: Van Der Waals EOS, RK: Redlich Kwong EOS, PR: Peng-Robinson EOS).

However, we find in the literature specific equations of state exactly representing the phase behaviour of pure substances. For \( \text{CO}_2 \), Bender (1970) gives a good presentation of
the PVT surface; the density can be correctly calculated in the critical region. As we will see later, in order to calculate all the thermodynamic quantities as well as the different terms of the Jacobian matrices of the Euler fluxes, we need to evaluate the derivates of the pressure with respect to the density, temperature, total energy, velocities, etc. Using the formulation proposed by Bender, the determination of these derivatives is not easy and manageable expressions can not be obtained.

Altunin and Gadetskii proposed a suitable analytical EOS for CO$_2$ (Altunin et al., 1971), which satisfactorily fits data given in the IUPAC’s stables (Angus et al., 1975). This EOS is written as follows:

\[
Z = 1 + \omega \sum_{i=0}^{9} \sum_{j=0}^{6} b_{ij}(\tau - 1)^i(\omega - 1)^j,
\]

where \( \tau = T_c/T \) and \( \omega = \rho/\rho_c \).

Here, \( T_c \) and \( \rho_c \) are the temperature and the density at the critical point, and \( Z \) the compressibility factor \( (P/\rho T) \). We notice that at low density, Eq. (10) coincides with the EOS of perfect gas \( (Z \approx 1) \).

A suitable EOS of the form \( P = P(\rho, T) \) allows us to determine the specific heats, the speed of sound, and some derivatives needed in the Riemann solver.

- The specific heat at constant volume \( C_v \) for a dense gas at a temperature \( T \) and density \( \rho \) can be calculated as

\[
C_{v,\rho} = C_{v_0} - \int_0^\rho \left( \frac{T}{\rho^2} \right) \left( \frac{\partial^2 P}{\partial T^2} \right) \frac{d\rho}{\rho}.
\]

Calculation of the first term \( C_{v_0} \) is based on the value of the fundamental frequencies of the molecules under consideration. Altunin gives an approximation of this term:

\[
C_{v_0} = \sum_{i=0}^{7} \gamma_i T^i - R,
\]

where \( R \) is the perfect gas constant \( (R=188.92 \text{ J/K.Kg for CO}_2) \).

- The heat capacity at constant pressure is related to the heat capacity at constant volume by the expression

\[
C_p = C_v + \frac{T}{\rho^2} \left( \frac{\partial P}{\partial \rho} \right)_T \frac{\partial}{\partial T} \frac{\partial P}{\partial \rho}.
\]

- The velocity of sound may be written as

\[
c^2 = \left( \frac{C_p}{C_v} \right) \left( \frac{\partial P}{\partial \rho} \right)_T.
\]

- The enthalpy is calculated as

\[
h = (h_{T}^i - h_{0}^i) + \Delta h_0 + \frac{P}{\rho} - RT + \int_0^\rho \left[ P - T \left( \frac{\partial P}{\partial T} \right) \right] d\rho,
\]
where \((h_T^2 - h_0^2)\) is the perfect gas value at temperature \(T\) when the zero reference state \((h_0^2)\) is taken to be the enthalpy of the ideal gas at \(T = 0\). The constant \(\Delta h_0\) is the enthalpy of sublimation at \(T = 0\), namely 26250 J/mol for Carbon Dioxide.

- We define the thermal isochoric factor as

\[
\alpha_v = \frac{P}{T} \left( \frac{\partial T}{\partial P} \right)_\rho.
\]

- The partial derivatives \(\left( \frac{\partial P}{\partial (\rho I)} \right)_\rho\) and \(\left( \frac{\partial P}{\partial \rho \rho I} \right)_\rho\) involved in the Riemann solver, can be calculated from the derivative tables of Borel (Borel, 1991), as follows:

\[
P_{\rho I} = \left( \frac{\partial P}{\partial (\rho I)} \right)_\rho = \frac{1}{\rho} \left( \frac{\partial P}{\partial I} \right)_\rho = \frac{P}{\alpha_v C_v \rho T}
\]

and

\[
P_\rho = \left( \frac{\partial P}{\partial \rho} \right)_I = c^2 + P_{\rho I} h,
\]

where, in the previous relationship, \(I\) refers to the internal energy.

Using the EOS (10), the different quantities \(C_p, C_v, \alpha_v\), etc., are easily obtained as functions of the temperature ratio \(\tau\), the density ratio \(\omega\), and the constant coefficients \(b_{ij}\) and \(\gamma_i\).

2.3. Discretization

In this study, it is useful to express the Navier-Stokes and energy equations in arbitrary curvilinear coordinates. Let \(\xi, \eta\) be the independent variables:

\[
\xi = \xi(x, y) \quad \eta = \eta(x, y).
\]

We define the Jacobian transformation \(J\) as

\[
J = \frac{\partial (\xi, \eta)}{\partial (x, y)} = \xi_x \eta_y - \xi_y \eta_x.
\]

Then the fully conservative form of the transformed Navier-Stokes equations can be written as

\[
\frac{1}{J} \frac{\partial \hat{Q}}{\partial t} + \frac{\partial \hat{F}}{\partial \xi} + \frac{\partial \hat{G}}{\partial \eta} + \frac{\hat{G}}{J, y} = 0,
\]

and

\[
\hat{Q} = Q
\]

\[
\hat{F} = (\xi_x F + \xi_y G) / J
\]

\[
\hat{G} = (\eta_x F + \eta_y G) / J
\]

The first-order derivatives included in \(F_v\), and \(G_v\) can be transformed in a similar manner.
2.3.1. Jacobian matrix, eigenvalues and eigenvectors

Many solvers need the eigenvalues and eigenvectors of the Jacobian matrix of the Euler fluxes. In our case, the Jacobian matrix is defined in the two directions by:

\[ A = \partial F / \partial Q \]
\[ B = \partial G / \partial Q. \]

In the curvilinear system \((\xi, \eta)\), the Jacobian matrices \(\tilde{A}\) and \(\tilde{B}\) are related to \(A\) and \(B\) as follows:

\[ \tilde{A} = \frac{(\xi_x A + \xi_y B)}{J} = Ja(\xi_x / J, \xi_y / J) \]
\[ \tilde{B} = \frac{(\eta_x A + \eta_y B)}{J} = Ja(\eta_x / J, \eta_y / J). \]

These matrices can be written in a general form using \(Ja(\alpha, \beta)\) where \(\alpha\) and \(\beta\) represent the coordinate transformation in each direction:

\[
Ja(\alpha, \beta) = \begin{pmatrix} 0 & \frac{\alpha}{\sqrt{\alpha^2 + \beta^2}} & \frac{\alpha}{\sqrt{\alpha^2 + \beta^2}} \\
\frac{\alpha}{\sqrt{\alpha^2 + \beta^2}} & 0 & \frac{\beta v}{\sqrt{\alpha^2 + \beta^2}} \\
\frac{\alpha}{\sqrt{\alpha^2 + \beta^2}} & \frac{\beta v}{\sqrt{\alpha^2 + \beta^2}} & 0 \\
\frac{\alpha v}{\sqrt{\alpha^2 + \beta^2}} & \frac{\beta u - \alpha v P_{pl}}{\sqrt{\alpha^2 + \beta^2}} & \frac{\alpha P_{pl}}{\sqrt{\alpha^2 + \beta^2}} \\
\frac{\alpha v}{\sqrt{\alpha^2 + \beta^2}} & \frac{\beta u + \alpha v P_{pl}}{\sqrt{\alpha^2 + \beta^2}} & \frac{\beta P_{pl}}{\sqrt{\alpha^2 + \beta^2}} \\
\frac{\beta u - \alpha v P_{pl}}{\sqrt{\alpha^2 + \beta^2}} & \frac{\beta u + \alpha v P_{pl}}{\sqrt{\alpha^2 + \beta^2}} & 0 \\
\end{pmatrix}
\]

Here \(q^2/2\) is the kinetic energy \(\frac{u^2 + v^2}{2}\) and \(H = h + \frac{q^2}{2}\) is the total enthalpy. The derivative \(P_{pl}\) is given by (17).

The four eigenvalues of \(\tilde{A}\) and \(\tilde{B}\) are calculated as

\[ \lambda_1 = \alpha u + \beta v \]
\[ \lambda_2 = \alpha v + \beta u \]
\[ \lambda_3 = \alpha v + \beta u + c \sqrt{\alpha^2 + \beta^2} \]
\[ \lambda_4 = \alpha u + \beta v - c \sqrt{\alpha^2 + \beta^2}. \]

The Jacobian matrix \(Ja(\alpha, \beta)\) can be written as

\[ Ja = R\Lambda R^{-1}, \]

where \(\Lambda\) is a diagonal matrix whose elements are the eigenvalues of \(Ja\), which are the characteristic speeds in the direction \((\alpha, \beta)\).

The corresponding right-eigenvectors matrix is

\[
R = \begin{pmatrix} \frac{1}{c^2} & \frac{1}{c^2} & \frac{1}{c^2} \\
\frac{u}{c^2} & \frac{u + ac}{c^2} & \frac{u - ac}{c^2} \\
\frac{v}{c^2} & \frac{v + bc}{c^2} & \frac{v - bc}{c^2} \\
\frac{1}{c^2} \left( H - \frac{c^2}{P_{pl}} \right) & \frac{-bu + av}{c} & \frac{H + (au + bv)c}{c^2} & \frac{H - (au + bv)c}{c^2} \\
\end{pmatrix}
\]
and its inverse matrix can be written as

\begin{equation}
R^{-1} = \begin{pmatrix}
P_{pl}(H - q^2) & P_{pl}u & P_{pl}v & -P_{pl} \\
(bu - av)c & -bc & ac & 0 \\
\frac{P_{pl}(H - q^2) + c^2 + c(au + bv)}{2} & \frac{(P_{pl}u - ac)}{2} & \frac{(P_{pl}v - bc)}{2} & \frac{P_{pl}}{2} \\
\frac{P_{pl}(H - q^2) + c^2 + c(au + bv)}{2} & \frac{(P_{pl}u + ac)}{2} & \frac{(P_{pl}v + bc)}{2} & \frac{P_{pl}}{2}
\end{pmatrix},
\end{equation}

where

\begin{equation}
\begin{align*}
a &= \frac{\alpha}{\sqrt{\alpha^2 + \beta^2}} \\
b &= \frac{\beta}{\sqrt{\alpha^2 + \beta^2}}.
\end{align*}
\end{equation}

2.3.2. Riemann solver

The generalisation of the Roe averaging technique (Roe, 1981) for a real gas has been carried out by Glaister (1988) or Montagné et al. (1987). The flux at a point separating two states \( Q^L \) and \( Q^R \) is based on the eigenvalues and eigenvectors of some average of \( \overline{A} \) and \( \overline{B} \). The optimum choice of these matrices is one satisfying:

\begin{equation}
\Delta \overline{F} = \overline{A} \Delta (\overline{Q}) \\
\Delta \overline{\phi} = \overline{B} \Delta (\overline{Q}),
\end{equation}

where \( \Delta \) is defined by:

\begin{equation}
\Delta(\cdot) = (\cdot)^R - (\cdot)^L.
\end{equation}

The choice of \( \overline{A} \) and \( \overline{B} \) will capture discontinuities exactly; to obtain these matrices, we can seek for an average state \( \overline{Q} \), such that

\begin{equation}
\overline{A} = \overline{A}(\overline{Q})
\end{equation}

and

\begin{equation}
\overline{B} = \overline{B}(\overline{Q}).
\end{equation}

Such a state is known as the Roe-averaged state.

The parameters of \( \overline{A} \) and \( \overline{B} \) depend explicitly on the thermodynamic variables, \( H, P_{pl} \) and \( c \). Since the density is not explicitly required, one would expect the Roe-averaged
state to depend only on $\rho^L$ and $\rho^R$ through their ratio. Therefore, we define $\delta$ as

$$\delta = \sqrt{\frac{\rho^R}{\rho^L}}.$$  

(32)

The average density which refers to the first component of $\tilde{Q}$ is written as

$$\tilde{\rho} = \sqrt{\rho^R \rho^L}.$$  

(33)

In order to express the averaged quantities in $\tilde{A}$ and $\tilde{B}$, we first need the average of the velocity components, the total and specific enthalpies complying with the Roe averaging conditions given in Roe (1981):

$$\tilde{u} = \frac{u^L + \delta u^R}{1 + \delta}, \quad \tilde{v} = \frac{v^L + \delta v^R}{1 + \delta}, \quad \tilde{H} = \frac{H^L + \delta H^R}{1 + \delta}.$$  

(34)

We note that these quantities do not depend on the form of the equation of state and can be written similarly for a perfect gas flow.

Using Eq (34), we define the Roe-averaged specific enthalpy $\tilde{h}$ as

$$\tilde{h} = \tilde{H} - \frac{\tilde{\rho}^2}{2},$$  

(35)

that is

$$\tilde{h} = \frac{\tilde{h}^R + \delta \tilde{h}^L}{1 + \delta} + \frac{\delta}{(1 + \delta)^2} \left( (\Delta u)^2 + (\Delta v)^2 \right).$$  

(36)

Regarding the sound speed, the estimation of an average state depends on the EOS employed. In fact, the averaged value of $c$ is given by

$$\tilde{c}^2 = \frac{\tilde{P} + \tilde{P}_{\rho I}\tilde{h}}{\tilde{\rho}}.$$  

(37)

In order to estimate $\tilde{c}$ as well as the components of the eigenvectors matrices $R$ and $R^{-1}$, the averaged values of the pressure derivatives ($P_{\rho}$ and $P_{\rho I}$) must be evaluated.

For a perfect gas, $P_{\rho} = 0$ and $P_{\rho I} = (\gamma - 1), (\gamma$ is the heat capacity ratio); however, for a real gas (dense gas), Montagné et al. (1987) show that the average values of $\tilde{P}_{\rho I}$ and $\tilde{P}_{\rho}$ must depend on the thermodynamic states on the right ($R$) and left ($L$) sides and not on the Roe average value of the specific enthalpy ($h$) as for a perfect gas. According to Montagné, $\tilde{P}_{\rho I}$ and $\tilde{P}_{\rho}$ are calculated using the expressions:

If $\Delta P \neq 0$ and $\Delta \rho \neq 0$ then

$$\frac{1}{\tilde{P}_{\rho I}} = \frac{1}{2} \left[ \left( \frac{P_{\rho I}^L}{\tilde{P}_{\rho I}} + \frac{P_{\rho I}^R}{\tilde{P}_{\rho I}} \right) \frac{\Delta P}{\Delta (\rho I) \Delta P} + \left( \frac{1}{P_{\rho I}^L} + \frac{1}{P_{\rho I}^R} \right) \frac{\tilde{c}^4 \Delta \rho}{\Delta P} \right] \Delta (\rho I).$$  

(38)
\[
\bar{P}_\rho = \bar{P}_{\rho L} - \frac{1}{2} \left[ \left( \frac{P^L_{\rho L}}{P^L_{\rho L}} + \frac{P^R_{\rho L}}{P^R_{\rho L}} \right) \Delta P + \left( \frac{1}{P^L_{\rho L}} + \frac{1}{P^R_{\rho L}} \right) c^2 \Delta \rho \right] \frac{\Delta P - c^2 \Delta \rho \Delta (\rho I)}{\Delta P^2 + c^2 \Delta \rho^2}
\]

If \( \Delta P = 0 \) and \( \Delta \rho = 0 \) then \( \bar{P}_{\rho L} = \frac{P^L_{\rho L} + P^R_{\rho L}}{2} \), \( \bar{P}_\rho = \frac{P^L_{\rho L} + P^R_{\rho L}}{2} \).

To ensure that these average values are unique at the interface between states R and L, Montagné et al. use a scale factor \( c \) which is proportional to the ratio \( \Delta P / \Delta \rho \):

\[
c^2 = \frac{(c^L)^2 + (c^R)^2}{2}.
\]

Montagné et al. showed that the formulations (38, 39) can be used when the equation of state is a nonconvex function (generally the case for the real gas equation of state). These formulae need only the different values in both right and left states. For this reason, this model is chosen and the numerical approach of the averaged pressure derivatives is used in our calculation. The quantities \( P^R_{\rho L}, P^L_{\rho L}, P^R_{\rho R} \) and \( P^L_{\rho R} \) can be calculated from a combination of Eqs. (14), (17) and (18).

As reported by Glaister (1988), \( \Delta (\rho I) \) can be written in different forms, but the best known and natural choice is:

\[
\Delta (\rho I) = \bar{\rho} \Delta I + \bar{I} \Delta \rho.
\]

In addition, this formula is consistent with the expressions for the pressure derivatives given in (38 and 39).

Finally, the averaged internal energy is defined by

\[
\bar{I} = \frac{I^L + \delta I^R}{1 + \delta}.
\]

These mean quantities allow us to calculate the components of the Jacobian matrices \( \bar{A} \) and \( \bar{B} \). Numerically, we check the value of \( \bar{c} \) to be sure that it lies inside the range given by \( c^L \) and \( c^R \).

2.3.3. Scheme and algorithm

The integration of the governing Eqs. (19) is performed by means of a shock capturing finite-difference method based on an implicit symmetric TVD scheme (Harten, 1986 and Yee, 1987).

If we consider a grid point where \( \xi = i \Delta \xi \) and \( \eta = j \Delta \eta \) and the quantities at time \( t = n \Delta t \), the discretization of Eq. (20) is written below:

\[
\begin{align*}
\bar{\bar{Q}}_{i,j}^{n+1} - \bar{Q}_{i,j}^n & + \lambda^\xi \theta \left[ \bar{F}_{i+1/2,j}^{n+1} - \bar{F}_{i-1/2,j}^{n+1} \right] + \lambda^\eta \theta \left[ \bar{G}_{i,j+1/2}^{n+1} - \bar{G}_{i,j-1/2}^{n+1} \right] \\
& = -\lambda^\xi (1-\theta) \left[ \bar{F}_{i+1/2,j}^n - \bar{F}_{i-1/2,j}^n \right] - \lambda^\eta (1-\theta) \left[ \bar{G}_{i,j+1/2}^n - \bar{G}_{i,j-1/2}^n \right] - \Delta \xi \Delta \eta \Delta t \frac{\bar{\bar{Q}}_{i,j}^n}{y_{i,j}},
\end{align*}
\]
with $\lambda_\xi = \frac{\Delta t}{\Delta x}$ and $\lambda_\eta = \frac{\Delta t}{\Delta y}$.

As we look for a steady state solution, the fully implicit scheme is only first-order accurate in time ($\theta = 1$). The discretization of the diffusive fluxes ($F_\nu, G_\nu$) is obtained through the use of a classic second-order central differencing approximation.

At the mid point of each cell, we consider a local system of characteristic fields and the scheme is applied in each characteristic direction (Yee, 1987).

Following Harten and Yee, the Euler flux function $\hat{F}$ in the $\xi$ direction is written:

$$\hat{F}_{i+1/2,j} = \frac{1}{2} \left[ \hat{F}_{i,j} + \hat{F}_{i+1,j} + R_{i+1/2} \Phi_{i+1/2} \right],$$

where $R$ is the column eigenvector matrix. It is calculated with the Roe averaging technique and the elements are defined previously in matrix form (25).

The diffusive part of this numerical flux $\Phi_{i+1/2}$ is a matrix with its elements denoted by $\varphi_{i+1/2}^l$. In order to ensure that this scheme is a second-order TVD scheme, the $\Phi$ elements must be written:

$$\varphi_{i+1/2}^l = \psi \left( a_{i+1/2}^l \right) \left[ \alpha_{i+1/2}^l - \Xi_{i+1/2}^l \right] \quad l = 1, ..., 4.$$

$\alpha_{i+1/2}^l$ stands for the forward difference of the local characteristic variables in the $\xi$ direction and is defined below:

$$\alpha_{i+1/2}^l = R_{i+1/2}^{-1} \left( \tilde{Q}_{i+1,j} - \tilde{Q}_{i,j} \right)$$

The value $a_{i+1/2}^l$ is the characteristic speed in the $\xi$ direction for the Jacobian matrix evaluated at the Roe average between $Q_{ij}$ and $Q_{i+1,j}$.

The function $\psi(z)$ is a Lipschitz continuous function used to prevent zero dissipation in the vicinity of rarefaction wave; it is defined as

$$\psi(z) = |z| \quad \text{if} \quad |z| \geq \varepsilon$$

$$\psi(z) = \frac{|z^2 + \varepsilon^2|}{2\varepsilon} \quad \text{if} \quad |z| < \varepsilon,$$

where $\varepsilon$ is the entropic dissipation parameter which is normalised by the spectral radius $\rho(A_\xi)$ of the Jacobian matrix of the conservative flux in the $\xi$ direction:

$$\varepsilon = \varepsilon_1 \rho(A_\xi).$$

Here $\varepsilon_1$ is a small positive number. In the following calculations $\varepsilon_1$ is constant and equal to $10^{-3}$.

Finally $\Xi_{i+1/2}^l$ is a limiter function; it acts as an anti-diffusive flux. The limitation is performed using the minmod function; it can be expressed as

$$\Xi_{i+1/2}^l = \text{minmod} \left( \alpha_{i-1/2}^l, \alpha_{i+1/2}^l, \alpha_{i+3/2}^l \right).$$
Function minmod is defined as

\[
\text{minmod}(x, y, z) = \begin{cases} 
\min(|x|, |y|, |z|) & \text{if } \text{Sign}(x) = \text{Sign}(y) = \text{Sign}(z) \\
0 & \text{Otherwise}
\end{cases}
\]

It is well known that this limiter lies in the classic second-order TVD limiter region (Waterson and Deconinck, 1995). Similar formulations could be written in the $\eta$ direction.

2.3.4. Formulation ADI

To solve $\tilde{Q}^{n+1}$ in (42) one normally needs to solve a set of non-linear algebraic equations iteratively with standard methods. One way to avoid this is to linearize the implicit operator and solve the linearized form by a conservative linearized ADI. For equation (42), the ADI form can be written as

\[
\left[ I + \lambda^\xi U^\xi_{i+1/2,j} - \lambda^\xi U^\xi_{i-1/2,j} \right] D^* = -\lambda^\xi \left[ \tilde{P}^n_{i+1/2,j} - \tilde{P}^n_{i-1/2,j} \right] - \lambda^\eta \left[ \tilde{G}^n_{i+1/2,j} - \tilde{G}^n_{i-1/2,j} \right] - \Delta t \left( \frac{C^n}{y} \right)_{i,j} \Delta \xi \Delta \eta.
\]

Then

\[
\left[ I + \lambda^\eta U^\eta_{i,j+1/2} - \lambda^\eta U^\eta_{i,j-1/2} \right] D^n = D^*
\]

and finally

\[
\tilde{Q}^{n+1} = \tilde{Q}^n + D^n.
\]

Here $U$ is expressed as

\[
U^\xi_{i+1/2,j} = \frac{1}{2} \left[ \tilde{A}_{i+1,j} - \Omega^\xi_{i+1/2,j} \right]
\]

and

\[
U^\eta_{i,j+1/2} = \frac{1}{2} \left[ \tilde{B}_{i+1,j} - \Omega^\eta_{i,j+1/2} \right].
\]

As we look for a steady state solution, the scheme needs to be of second-order accuracy in space. Therefore, according to Yee and Harten, the matrices $\Omega^l_{i,j}$ are written as:

\[
\Omega^l_{i,j} = R_{i,j} \text{diag}\left( \psi(a^l) \right) R^{-1}_{i,j}.
\]

2.3.5. Boundary conditions

The grid domain is composed of two parts (Fig. 3), a narrow cylindrical nozzle and a rectangular expansion domain whose dimensions are larger than the nozzle’s. This domain is limited by a flat plate positioned normally to the jet axis.
Three types of boundary conditions can be defined, those at the wall/liquid interface, those at the inflow and outflow boundaries and those on the symmetry axis.

- In the first type (wall/liquid interface), we consider that
  - At the nozzle wall (Fig. 3), we prescribe the temperature \( T^{n+1} = T_{\text{nozzle}} \), and we impose no slip conditions \( u^{n+1} = 0, \ v^{n+1} = 0 \).
  - At the downstream boundary of the computational domain (Fig. 3), we impose that there exists an adiabatic flat plate \( \frac{\partial T}{\partial x} = 0 \) and we also impose a no slip boundary condition \( u^{n+1} = 0, \ v^{n+1} = 0 \).
- In the second type we suppose that
  - At the inflow (Fig. 3), quantities \( (u^0, v^0, E^0, T_{\text{nozzle}}) \) are assigned.
  - At the outflow boundary (Fig. 3), we suppose that there exists no variation in the direction normal to the axis \( \frac{\partial (\rho u)}{\partial y} = 0 \).
- Finally, on the symmetry axis, we solve the governing equations setting \( (v = 0) \) on this line. We remark that on the axis, \( u, \ P, \ T, \ \rho, \) and \( E \) are symmetric quantities but \( v \) is anti-symmetric.

3. Numerical results

In order to check the TVD scheme expressed above and the different choices made in writing this scheme, we first applied the algorithm, written for the one-dimensional Euler equations, to the calculation of the shock tube problem described by Sod (1978). A diaphragm, positioned at \( x/L = 0.5 \) (where \( L \) is the tube length and \( x = 0 \) refers to the left end of the tube) initially separates two regions with constant states. On the left side of the diaphragm, the density and the pressure are prescribed at supercritical values \( (\rho_{\infty} = 470 \text{ mg.m}^{-3} \text{ and } P_{\infty} = 1.54 \times 10^7 \text{ Pa}) \). The density and pressure ratios between the left and right parts are respectively \( \rho_{\infty}/\rho_{\text{out}} = 8 \) and \( P_{\infty}/P_{\text{out}} = 10 \). On both sides of the diaphragm, the fluids are initially at rest. The calculation is performed using...
200 points along the tube. The time step is prescribed, corresponding to a maximum CFL number close to 0.2. At the initial state, the diaphragm is broken and three waves occur in the tube: a rarefaction wave moving to the left, a contact discontinuity and a shock wave moving to the right; these waves separate different uniform flow regions as we can see on the distribution of the density along the tube (solid line in Figure 4). It shows also the distribution of the velocity (dashed line) and pressure (dotted line); these quantities exhibit important variations in the rarefraction and the shock waves while they stay constant through the contact discontinuity. These results can be compared with those obtained by Sod and Roe for instance and we must mention that good agreement is obtained even for a dense gas.

Fig. 4. – Distribution along a shock tube of the density, the velocity and the pressure resulting from a one-dimensional calculation: \( \rho_\infty = 470 \text{ kg.m}^{-3} \); \( P_\infty = 1.54 \times 10^7 \text{ Pa} \); \( \rho_\infty / \rho_{\text{out}} = 8 \), \( P_\infty / P_{\text{out}} = 10 \).

The calculation of the expanded jet of supercritical carbon dioxide downstream a cylindrical throttle was then undertaken. The resolution of the governing equations give us the values of the non-conservative quantities (\( \rho, u, v, E \)). The temperature and the pressure must be calculated by the EOS (10). Since this former is highly non linear, a root finding technique must be employed to know the values of \( T \) and \( P \). So, using Eqs. (10) and (15), the temperature can be expressed as a non linear function of the density, of the total energy and of the velocity components. A Newton method is used then to calculate the temperature. The pressure is therefore deduced from (10) knowing the density (\( \rho \)) and the temperature (\( T \)). The convergence in the Newton procedure, judged by the difference in temperature between two successive iterations (using an accuracy equal to \( 10^{-10} \)) is reached often less than 7 Newton steps.
The present calculations are performed using a constant Courant number equal to CFL=5. Therefore, the time step is calculated locally, using this CFL number and the spectral radius of the Jacobian matrices of the Euler fluxes in both directions.

The numerical code was checked in the calculation of the expanded jet of supercritical carbon dioxide downstream of a cylindrical throttle maintained at a constant temperature. Three calculations are performed depending on the upstream and the downstream flow conditions.

The first calculation presented is called the subsonic case, because the Mach number at the inlet is equal to 0.58. At the upstream boundary, the carbon dioxide is in a supercritical state since the values of the pressure \( P \) and the temperature \( T \) are greater than the values of the corresponding quantities at the critical point \( (T_c=304.21 \text{ K, } P_c=73.78 \times 10^5 \text{ Pa}) \):

\[
T_\infty = 388 \text{ K}; \quad P_\infty = 236.05 \times 10^5 \text{ Pa} \\
\rho_\infty = 500 \text{ kg.m}^{-3}; \quad u_\infty = 200 \text{ m.s}^{-1}
\]

In the expansion domain, (Fig. 3), at the initial state, the thermodynamic conditions are \( T_{\text{ex}} = 353 \text{ K}; P_{\text{ex}} = 1.114 \times 10^6 \text{ Pa}; \rho_{\text{ex}} = 1.720 \text{ kg.m}^{-3} \). These conditions correspond to the state of Carbon Dioxide gas without motion \( (u_{\text{ex}} = v_{\text{ex}} = 0 \text{ m.s}^{-1}) \). These conditions lead to a pressure ratio between the inlet and the expansion chamber equal to \( P_\infty / P_{\text{ex}} = 210 \). The temperature on the flat plate at the downstream boundary is set initially at the temperature of the fluid in the expansion domain. The capillary is \( 5 \times 10^{-3} \text{ m} \) long and its diameter is \( D = 330 \times 10^{-6} \text{ m} \). The flat plate is located at about 90 capillary diameters downstream from the outlet of the device. The typical mesh size used for this calculation is 60 points in the streamwise direction and 80 points in the direction normal to the axis.

The convergence of the present calculation is judged on the evolution of the \( L_2 \) norm of the residual. In Figures 5 and 6 we can see the evolution of the \( L_2 \) norm of both

\[
\text{Log}_{10} |\rho|_{L_2}
\]

Fig. 5. – Evolution of the \( L_2 \) norm of the density residual versus number of iterations.
the density and the axial momentum versus the number of iterations; the magnitude of the residual dropped tremendously in 3000 steps and the convergence toward the steady state solution is obtained after about 9000 iterations.

In Figures 7, 8, 9 we present the evolution of the dimensionless pressure, density and temperature along the axis. The rapid expansion is located just downstream of the outlet of the capillary (x=0, refers to the outlet of the capillary). The critical values of $P$, $\rho$, $T$ (dashed line respectively in Figures 7, 8, 9), are reached at the outlet, which means that the fluid rapidly losses its solvent power. In other words, if the supercritical fluid is saturated with dissolved solid substances upstream of the capillary, these substances will precipitate into solid particles very closed to the outlet of the nozzle.

It is important for the fluid to expand from the supercritical phase to the gas phase without crossing the liquid phase. If the liquid state is reached, there exists a condensation region in the computational domain. Of course, in that region, the fluid would be quasi-incompressible and using our code, we should have no chance of obtaining a good

---

Fig. 6. – Evolution of the $L_2$ norm of the longitudinal momentum residual versus number of iterations.

Fig. 7. – Longitudinal evolution of the static pressure ($P/P_\infty$) along the axis: $U_\infty = 200$ m.s$^{-1}$, $P_\infty = 236.05$ $10^5$ Pa, $T_\infty = 388$ K, $P_{\infty}/P_{cr} = 210$.
representation of the flow. We also mention that, in the expansion, the path of the thermodynamic variables does not cross the critical region (CPLV – Fig. 1) where there are physical discontinuities of several thermodynamic properties (compressibility, viscosity... tend to infinity).

This expansion leads to a sudden spreading of the jet in the motionless fluid as we can see on Figures 10, 11 and a great acceleration of the flow is seen, especially along the axis. (In order to identify the fluid motion clearly, an enlargement, magnitude 2, is applied in the radial direction). When we look at the velocity field further downstream of this accelerated region, we see a first recompression zone where a small Mach disk takes place (Fig. 10). Successive zones of expansion and recompression are then visible, mainly on the isovalues of the longitudinal velocity ($U$) (Fig. 10). The position of the Mach disk as well as the location of the compression and expansion regions are clearly identified on the enlargement of the longitudinal evolution of the static pressure ($P$) along the axis (Fig. 7). The radial velocity ($V$), (Fig. 11), becomes alternatively positive (compression) and negative (expansion). The impinging jet diverges on the normal flat plate and a radial
boundary layer is then created along this plate. This boundary layer is first accelerated, up to a distance from the axis equal to 15-20 D, and then decelerated Fig. 11.

The growth of solid particles is sensitive to the temperature. As we can see in Figure 12, the main core of the jet is at very low temperature; a significant temperature gradient takes place across the jet as well as along the plate (Fig. 13) since we consider an adiabatic plate. The minimum value of the temperature on the plate is situated at 2 diameters from the axis, Fig. 13 (and not on the axis). This might be explained by the fact that, close to the plate between the axis and \( y = -2.0 \), there exists a dead-water region (\( v \) is positive and \( u \) exhibits very small values). If we suppose that small particles (light particles) are generated close to the outlet of the capillary in the vicinity of the axis, these particles will follow trajectories very close to the streamlines and will stay in the vicinity of the centerline of the flow. These light particles will be transported within a cold flow. On
the other hand, if we suppose that heavy particles are generated close to the outlet of the capillary, they will tend to move away from the axis due to the expansion of the jet and they will be subject to a positive temperature gradient across the jet.

The second test case consists in the simulation of the expansion jet into a low pressure chamber. The computational domain has the same dimensions and the same number of grid points than in the first calculation. The flow conditions at the inlet of the capillary are unchanged compared with the first test case. In the expansion chamber, the CO$_2$ gas is initially without motion ($u_{ex} = v_{ex} = 0$ m.s$^{-1}$). The initial conditions have been set to:

$$ T_{ex} = 353 \text{ K}, \quad P_{ex} = 0.666 \times 10^4 \text{ Pa}, \quad \rho_{ex} = 0.001 \text{ kg m}^{-3}. $$

Obviously, due to the very great pressure ratio between the inlet and the outlet, $P_{\infty}/P_{ex} = 354.10^3$, the jet spreads out abruptly downstream of the nozzle, as we can see for the isovalues of $U$ and $V$ (Figs. 14, 15). As in the first case, the critical values

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Fig. 14 – Isovalues of the longitudinal velocity \((U/U_\infty)\) for the second test case:
\[ U_\infty = 200 \text{ m}\cdot\text{s}^{-1}, \quad P_\infty = 236.05 \times 10^5 \text{ Pa}, \quad T_\infty = 388 \text{ K}, \quad P_\infty/P_{cE} = 354 \times 10^3. \]

Fig. 15 – Isovalues of the radial velocity \((V/U_\infty)\) for the second test case:
\[ U_\infty = 200 \text{ m}\cdot\text{s}^{-1}, \quad P_\infty = 236.05 \times 10^5 \text{ Pa}, \quad T_\infty = 388 \text{ K}, \quad P_\infty/P_{cE} = 354 \times 10^3. \]

of pressure, density and temperature are reached at the outlet of the capillary Fig. 16, 17, 18. Therefore, the nucleation and the condensation of the solute will occur out of the nozzle. It is important to notice that there is no recompression zone in the domain, except close to the normal flat plate where a sudden recompression is seen (see the enlargement on Fig. 16). The flow accelerates along the axis up to the vicinity on the flat plate where a boundary layer, exists (Fig. 19).

On the axis, as the kinetic energy has a relatively high value upstream of the plate, the sudden deceleration induces an important increase of the temperature. This is the reason
Fig. 16 – Longitudinal evolution of the static pressure \( (P/P_\infty) \) along the axis:
\[ U_\infty = 200 \text{ m.s}^{-1}, \quad P_\infty = 236.05 \times 10^5 \text{ Pa}, \quad T_\infty = 388 \text{ K}, \quad P_\infty / P_{\infty} = 354 \times 10^3. \]

Fig. 17 – Longitudinal evolution of the density \( (\rho/\rho_\infty) \) along the axis:
\[ U_\infty = 200 \text{ m.s}^{-1}, \quad P_\infty = 236.05 \times 10^5 \text{ Pa}, \quad T_\infty = 388 \text{ K}, \quad P_\infty / P_{\infty} = 354 \times 10^3. \]

Fig. 18 – Longitudinal evolution of the temperature \( (T/T_\infty) \) along the axis:
\[ U_\infty = 200 \text{ m.s}^{-1}, \quad P_\infty = 236.05 \times 10^5 \text{ Pa}, \quad T_\infty = 388 \text{ K}, \quad P_\infty / P_{\infty} = 354 \times 10^3. \]
why the temperature of the impinging point is relatively high and an important positive longitudinal gradient is located in the vicinity of the normal plate (Fig. 18). The radial gradient of the temperature along the plate (Fig. 20) is consistent with the variation of the radial velocity (V) in the vicinity of this plate (Fig. 15). The plate is heated by the flow (Figs 20, 21) the maximum value of the temperature is located at about 60 diameters away from the axis (Figs 20, 21) where the radial velocity is maximum (Fig. 15). Finally, let us mention that the maximum value of the static pressure on the normal plate is located at about 40 capillary diameters away from the axis (Fig. 22). This location coincides with the minimum value of the temperature (Fig. 20). This might be explained by an important curvature of the streamlines close to the plate and might be compared with the flow in a curved duct. In contrast, with low pressure ratio (case 1), the maximum pressure value on the plate is situated on the axis (Fig. 23). In fact, close to the axis, the streamlines seem to have less important curvature than for the large pressure ratio.

Finally, the third calculation deals with the supersonic jet. The Mach number at the inlet of the cylindrical capillary is equal to 1.45; compared to the first case, the longitudinal
Fig. 21 – Isovalues of the static temperature ($T/T_\infty$) for the second test case:
$U_\infty = 200 \text{ m.s}^{-1}$, $P_\infty = 236.05 \text{ 10}^5 \text{ Pa}$, $T_\infty = 388 \text{ K}$, $P_\infty/P_{ex} = 354 \text{ 10}^3$.

Fig. 22 – Evolution of the pressure ($P/P_\infty$) along the normal plate:
$U_\infty = 200 \text{ m.s}^{-1}$, $P_\infty = 236.05 \text{ 10}^5 \text{ Pa}$, $T_\infty = 388 \text{ K}$, $P_\infty/P_{ex} = 354 \text{ 10}^3$.

Fig. 23 – Evolution of the pressure ($P/P_\infty$) along the normal plate:
$U_\infty = 200 \text{ m.s}^{-1}$, $P_\infty = 236.05 \text{ 10}^5 \text{ Pa}$, $T_\infty = 388 \text{ K}$, $P_\infty/P_{ex} = 210$.  

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velocity \((u)\) is increased up to 500 m/s. The other thermodynamic quantities have the same values as in the first calculation, however, the length of the cylindrical nozzle is changed \((1.10^{-2} \text{ m})\). The flat plate is located at about 80 diameters downstream of the exit of the capillary. A finer grid than in the previous calculations \((100 \times 100)\) is used in this simulation. It is important to note that the critical values of the pressure and the temperature (dashed lines in Figs. 24, 25) are reached downstream of the outlet of the capillary, contrary to the two previous calculations. Compared to the previous flow, the nucleation of particles will take place slightly downstream. The flow seems to behave as in the first calculation, except that the magnitude of the variations of the quantities (for instance \(P\) and \(T\)) along the axis are much greater than in the first case. We notice that the Mach disk pressure rise takes place over a larger extent than in the subsonic case. The succession of the expansion and compression zones are more apparent in the present calculation mainly in the isovalues of the radial velocity \((v)\) (Fig. 26) which exhibit alternatively positive and negative values.

**Fig. 24** – Longitudinal evolution of the static pressure \((P/P_{\infty})\) along the axis:

\[
U_{\infty} = 500 \text{ m.s}^{-1}, \quad P_{\infty} = 236.05 \times 10^5 \text{ Pa}, \quad T_{\infty} = 388 \text{ K}, \quad P_{\infty}/P_{\text{cr}} = 210.
\]

**Fig. 25** – Longitudinal evolution of the temperature \((T/T_{\infty})\) along the axis:

\[
U_{\infty} = 500 \text{ m.s}^{-1}, \quad P_{\infty} = 236.05 \times 10^5 \text{ Pa}, \quad T_{\infty} = 388 \text{ K}, \quad P_{\infty}/P_{\text{cr}} = 210.
\]
Fig. 26 – Isovalues of the radial velocity ($V/\bar{V}_\infty$) for the third test case:
$\bar{V}_\infty = 500 \text{ m.s}^{-1}$, $P_\infty = 236.05 \times 10^5 \text{ Pa}$, $T_\infty = 388 \text{ K}$, $P_\infty/P_{\text{ce}} = 210$.

Fig. 27 – Evolution of the temperature ($T/T_\infty$) along the normal plate:
$\bar{V}_\infty = 500 \text{ m.s}^{-1}$, $P_\infty = 236.05 \times 10^5 \text{ Pa}$, $T_\infty = 388 \text{ K}$, $P_\infty/P_{\text{ce}} = 210$.

Fig. 28 – Density profile along the jet axis for two grid calculations.
As the kinetic energy of the fluid is more important than in the first case (Fig. 9), the temperature of the flat plate due to the deceleration is higher (Fig. 25). In Figure 27 the radial variation of the temperature along the normal plate is presented. While the plate is at a higher temperature than in the first calculation, the temperature behaves as in the subsonic case (compare Fig. 13 and Fig. 27).

Finally, we have tested the grid sensitivity, one subsonic case being calculated with two different grids (100 × 100) and (150 × 100). The calculations were performed for the two grid with the same dimensions and geometry for the nozzle and the expansion chamber. Figures 28 and 29 show the density and the vertical velocity profiles along the jet. We notice for the two calculations, the profiles in the nozzle are in agreement and calculations seem therefore to be independent of the grid. The same remark can be made for the main (first) expansion and near the plate. In contrast, the profiles along the jet have some sensitivity to the grid, and expansion and recompression zones are more amplified in the (150 × 100) grid.

![Diagram](image)

**Fig. 29** – Vertical velocity profile along the jet at a radius corresponding to the nozzle exit radius for two grid calculations.

4. Conclusion

The numerical simulation of the rapid expansion of supercritical fluid has been undertaken using the Navier-Stokes and total energy equations with a specific EOS proposed by Altinur and Gadestkii. This equation has been chosen since it is a good representation for the supercritical state of Carbon Dioxide compared with the IUPAC’s data, even very close to the critical region. The integration of these governing equations is performed by a finite-difference scheme through the use of Roe’s approximate Riemann solver written for a real gas. We have employed a symmetric TVD scheme based on the work of Harten and Yee using a minmod-like limiter function. The resolution of these equations is stabilised by an implicit step using the Alternating Direction Implicit (ADI) formulation. This algorithm allows us to employ a CFL number equal to 5. Using a
local time step, the scheme is second-order accurate in space; but, as we look for steady state solution, the scheme is only first-order accurate in time. To check the numerical code, three configurations (depending on the values of the inlet and the outlet conditions) have been investigated.

All the calculations are performed using approximately the same computational domain, and the CFL number is set equal to 5. In the first calculation, at the inlet of the nozzle, the flow is subsonic. The expansion takes place just at the outlet of the nozzle and the critical values of pressure, density, and temperature are reached “rapidly” just downstream of the capillary. A small Mach disk is well captured in this case. As the growth of the solid particles is sensitive to the temperature, we notice that the main core of the jet is at relatively low temperatures while an important positive gradient is recorded across the jet (in the radial direction).

The second calculation refers to the expansion of the jet into a very low pressure chamber. At the outlet of the nozzle, the jet diverges into the whole expansion domain. No recompression zone is visible along the jet axis, except very close to the normal flat plate where a rapid recompression takes place. This means that for the majority of the length of the centerline of the jet, the flow accelerates. Therefore, a very low temperature is recorded along this axis. An important radial gradient of the temperature is however created across the jet.

For the last calculation, the flow at the inlet of the capillary is supersonic instead of subsonic. This case is interesting since it presents a larger Mach disk than the first calculation. However, in general manner, while the longitudinal and the radial gradients of pressure and temperature are greater, the flow behaves as in the first calculation.

From a physical point of view, an abrupt decrease of the temperature within the jet allows the nucleation of particles. In this study, the influence of the flow conditions, at the inlet as well as at the outlet of the capillary, on the jet morphology has been reviewed. The size and distribution of solid particles along the normal plate is expected to depend strongly on the upstream and downstream boundary (or initial) conditions. Therefore, such jets provide an efficient mechanism to form solid materials in the form of powders or fibres.

In this study, we only studied the development of the jet downstream of a cylindrical capillary. It is reasonable to expect that the geometry of the capillary might have an important influence on the jet morphology and on the variation of the thermodynamic quantities; therefore, we are currently studying with the present numerical code, the evolution of the jet downstream of the capillary using different capillary shapes. The influence of mesh refinement seems to be minor. The Reynolds number (based on the capillary diameter) is high enough to assume that the jet flow is turbulent. In order to obtain more realistic simulations, investigations need to be undertaken to see the influence of the turbulence model on the flow geometry, using both mixing length schemes or classic $K - \varepsilon$ models. Finally, let us mention that simulations of nucleation and particle growth are being undertaken at the present time, the present numerical code providing the necessary fields of kinetic and thermodynamic quantities.
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