Chapter 16

Laser Assisted Aerodynamic Separation: An Overview

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16.1 Introduction

Among the many applications of lasers, one area that has been extensively researched over the last many years is in isotope enrichment, in particular, in utilizing it towards shrinking the size of the usually huge industrial enrichment plants. The reason behind this is the extraordinary purity of laser light that could be easily exploited to selectively excite the rarer isotope leading to its straightforward separation from the abundant species. Two approaches based on this simple concept viz., atomic vapor laser isotope separation (AVLIS) [165, 166] and molecular laser isotope separation (MLIS) [167] have emerged over the years although activity in MLIS, because of the prohibitively large laser intensity requirement, took a back-seat. Another laser-based approach that has gained momentum over the last decade or so basically combines isotope selective laser excitation with an existing separation scheme viz., aerodynamic separation [168] that takes advantage of the difference in the masses of the two isotopes. This laser assisted aerodynamic isotope separation scheme [169, 170] has established its clear advantage over the classical MLIS scheme in terms of power requirement from the laser.

It is well-known that an isotopic gaseous mixture upon expansion from an orifice will result in the heavier species tending to concentrate near the jet centerline while the lighter one moves outwardly [169] thus presenting the possibility of separating of the two species. The efficiency of the separation of isotopes here, understandably, depends on their relative mass difference and hence is unsuitable for heavier isotopes [171]. Further, polyatomic molecular gas seeded in an inert carrier gas subjected to supersonic nozzle expansion communicate with the low temperature bath provided by the monatomic carrier gas resulting in the relaxation of their various degrees of freedom, viz., the translational, rotational and the vibrational degrees of freedom [172, 173]. Thus, as the gas cools, the molecules in the jet form Van der Waals clusters with themselves (homo) and as well as with carrier atoms (hetero) [169, 170, 172, 173]. Selective laser excitation of one of the isotopic species is known to prevent its cluster formation thereby giving rise to a larger mass difference between the excited isotope and the non-resonant isotope that continue to form clusters, resulting in a much wider spatial separation between them [174, 175] thus making it compatible for enriching certain heavy isotopes of strategic interest [176–178]. Dilution of the molecular isotopic mixture in a buffer gas like Argon further enhances the overall separation efficiency as now the possibility of transferring the vibrational excitation energy from the resonant species to the non-resonant one through collisions reduces drastically [169, 170, 174]. Unlike the case of MLIS, where a single molecule needs to absorb tens of photons sequentially to reach the dissociation limit, the requirement on the intensity of the laser source is much less stringent here, thereby making this process accessible [175] to even continuous wave (CW) laser sources. For most molecules, CO_2 laser, that operates in the mid-IR region and tunable over 9-11 μm, although discretely, is the most attractive option to impart the selective excitation to the isotope of choice either directly or by way of converting its output into other wavelengths through non-linear or optical pumping routes. The first demonstration of dramatic enhancement of enrichment factor by this method made use of a 20 W CO_2 laser that selectively excited the abundant species, $^{32}SF_6$, in an expanding SF_6-Ar binary jet [169, 170]. Some important aspects of LANS process are,

- Supersonic expansion of process gas seeded in buffer gas.
- Knowledge of vibrational frequencies, isotope shift and selective excitation of the desired isotopic species when gas is sufficiently cooled (to have large population in the ground state) but before the onset of cluster formation.
- Skimming of the central stream without disturbing the expanding gas. Thus it is imperative to have a prior knowledge of various parameters associated with jet expansion to optimize the LANS process and the same are discussed in the following sections.

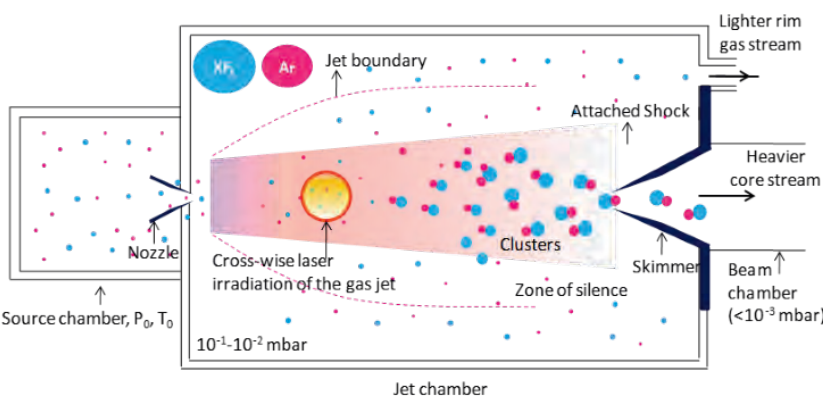

Figure 16.1: Pictorial representation of the three fundamental processes.

16.2 Supersonic Nozzle Expansion

Knowledge of various parameters viz., temperature, pressure, number density, velocity, Mach number etc., in the flow field of such supersonic jets is very crucial for increasing the separation efficiency in a LANS process. These flow properties can be accurately predicted using analytical expressions for isentropic fluid flow [179, 180]. To this end, various terms encountered in supersonic flow viz., compressible and incompressible fluid flow, Mach number, stagnation temperature, static temperature, dynamic temperature, stagnation pressure, static pressure, dynamic pressure, axial velocity, shocks etc., are discussed herein. Further, various properties of a continuum free-jet have been elaborated. The schematic of nozzle expansion from a source at (P_0, T_0) through a nozzle/orifice into an expansion chamber maintained at a background pressure P_b is shown in Fig. 16.1. The expansion being adiabatic, the velocity of the flow increases at the expense of internal energy of the fluid and hence the random thermal energy is converted to streamlined kinetic energy of the fluid flow resulting in the reduction of the local gas temperature. The substantial cooling associated with the isentropic expansion reduces the local sound speed in the expanding gas medium too, thereby increasing the Mach number in the supersonic flow. In the continuum region of the flow, collisions occur with sufficient rate for equilibrium to be maintained between the different degrees of freedom throughout the expansion process. However, at some point, the collision frequency in the expansion drops to such a value that a particular degree of freedom (translational, rotational or vibrational) may fall out of equilibrium. This is the beginning of transition from continuum to molecular flow regime that results in collision free environment and freezing of flow parameters [179, 180]. The rotational relaxation occurs more readily than vibrational, and, translational energy change occurs even more readily than rotational. Hence, $T_{tr} < T_{rot} < T_{vib}$ [179]. Thus, there exists a certain region in the expansion regime called the 'zone of silence' (Fig. 16.3) where the fluid properties are just right to exploit them for isotope selective studies, or, for that matter, for spectroscopy [179, 180].

16.3 Compressible and Incompressible Fluid Flow

It is well-known that sound propagates in a medium with speed that depends on the compressibility of the medium. The less compressible it is, the higher the speed of sound in it. Thus, speed of sound is a handy reference whenever a flow is involved. If the speed of the flow is much less compared to the speed of sound in the medium, the medium behaves as incompressible medium. However, if the flow velocity becomes comparable to the sound velocity, compressibility effects set in. The speed of sound itself can change from one point to another and so the velocity at each point has to be compared with the local sound speed for that point. This ratio is termed the Mach number and is defined as the speed at any point in the fluid (v) to the local sound speed (a), i.e.,

$$M = \frac{v}{a} \tag{16.1}$$

The medium is said to be compressible if there is a certain change in density, $\Delta\rho$, for a given change in pressure, ΔP. With a bit of mathematical juggle, a quantitative criterion to understand the compressibility effects in a fluid flow, is

$$\frac{\Delta\rho}{\rho} = M^2 \tag{16.2}$$

It can be seen that for $M < 0.3$, the change in density is less than 10%. This approximation is valid for incompressible flow. Hence compressibility effects become significant when Mach number exceeds 0.3, as is the case in supersonic expansion from a nozzle. Thus, fluid flow from a nozzle is treated as compressible flow.

16.4 Isentropic Relations of Compressible Fluid Flow

The isentropic relations for compressible fluid flow are as below:

$$\frac{T_0}{T} = 1 + \frac{\gamma - 1}{2}\frac{v^2}{a^2} \tag{16.3}$$

$$\frac{T_0}{T} = 1 + \frac{\gamma - 1}{2}M^2 \tag{16.4}$$

$$\frac{P_0}{P} = \left(\frac{T_0}{T}\right)^{\frac{\gamma}{\gamma-1}} \tag{16.5}$$

$$\frac{\rho_0}{\rho} = \frac{n_0}{n} = \left(\frac{T_0}{T}\right)^{\frac{1}{\gamma-1}} \tag{16.6}$$

Once M is known, all other thermodynamic parameters in the flow field can be evaluated.

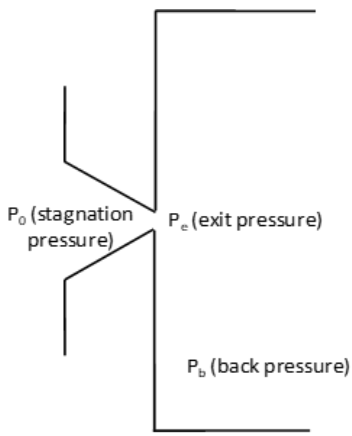

Figure 16.2: Expansion of gas from stagnation, P_0, into a background at P_b, P_e is the pressure at the nozzle exit.

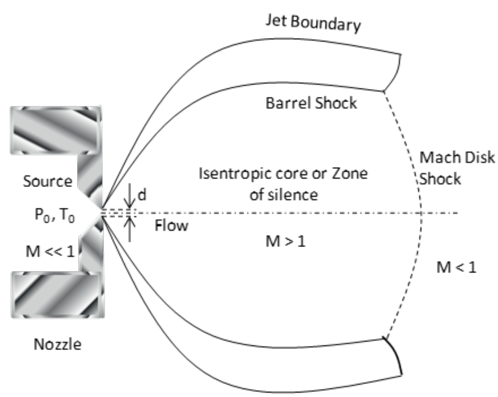

Figure 16.3: Continuum free-jet expansion from a nozzle at stagnation (P_0, T_0).

16.5 Free-jet Expansion

When the gas expands isentropically from a source at higher stagnation pressure into a chamber maintained at vacuum through an orifice such that the Knudsen number, $K_n = (\lambda/d)$ is $\ll 1$, i.e., $\lambda \ll$ d, where 'λ' is the mean free path (source chamber), 'd' is the characteristic dimension (here the nozzle diameter), the flow is referred to as free-jet as the gas expands free of the containing nozzle walls (Fig. 16.3). The gas expands from a negligibly small velocity inside the nozzle called the stagnation state (P_0, T_0) (M = 0) and with a pressure difference (P_0, P_b), and accelerates as the area decreases toward the nozzle exit. The flow may reach sonic speed (Mach number = 1), at the nozzle throat, provided the pressure ratio, P_0/P_b, equals a certain critical value, given by, $G = ((\gamma+1)/2)^{\frac{\gamma}{\gamma-1}}$, where γ, is the specific heat ratio. This value is less than 2.1 for all gases of practical interest [179]. If the pressure ratio is less than this G, the flow is subsonic with exit pressure 'P_e' equal to 'P_b', without any further expansion. As P_0/P_b increases beyond this critical ratio, the flow becomes sonic at the source exit and the exit pressure P_e = P* = $P_0\,(2/\gamma+1)^{\frac{\gamma}{\gamma-1}}$, independent of P_b. Since P_e now exceeds P_b, the flow is said to be under-expanded and a subsequent expansion occurs as the flow attempts to meet the necessary boundary condition imposed by the ambient pressure, P_b. i.e., $P_e = P_b$, for $P_b \geq P^*$, and $P_e = P^*$, for $P_b < P^*$ [Refer to Fig. 16.2]. Thus, a convergent nozzle accelerates subsonic flows with M < 1. For sufficient nozzle pressure ratio, the flow will reach sonic velocity at the nozzle throat. In this situation, Mach number equals 1 and the nozzle is said to be chocked. To be noted, increasing the pressure ratio further will not increase M beyond 1 at the throat, but downstream (i.e., external to the nozzle), the flow is free to expand supersonically (i.e., M > 1). Thus at the nozzle throat M = 1, and the the field parameters, T*, P* and ρ^* are referred to as chocked flow conditions. These parameters are obtained by substituting M = 1 in the Eqs. (16.3), (16.4) and (16.5), respectively and are derived to be,

$$\frac{T^*}{T_0} = \frac{2}{\gamma + 1} \tag{16.7}$$

$$\frac{P^*}{P_0} = \left(\frac{2}{\gamma + 1}\right)^{\frac{\gamma}{\gamma-1}} \tag{16.8}$$

$$\frac{\rho^*}{\rho_0} = \frac{n^*}{n_0} = \left(\frac{2}{\gamma + 1}\right)^{\frac{1}{\gamma-1}} \tag{16.9}$$

It is obvious from Eq. (16.8), that, in order to obtain supersonic free-jet expansion, the pressure ratio, P_o/P_b, should exceed $\left(\frac{\gamma+1}{2}\right)^{\frac{\gamma}{\gamma-1}}$ [179]. The maximum mass flow rate occurs at the throat of the nozzle given by, $\dot{m} = D.C.\rho^* A^* v^*$, where D.C. is the discharge coefficient (defined as the ratio of the actual discharge to the theoretical discharge) at the nozzle exit which is typically ~0.82 for gases of practical interest [181]. Supersonic flow has two crucial characteristics that make the expansion interesting. Firstly, in a supersonic flow, as the flow area increases, the velocity too increases, so that M becomes greater than 1, beyond the exit. This feature is much unlike subsonic flow where the velocity decreases as area increases. Secondly, a supersonic flow cannot sense downstream conditions. This is critical feature caused due to the fact that the information about the flow gets propagated at the speed of sound whereas fluid moves much faster i.e., M > 1. Thus, the flow does not know about the existing boundary condition, (the P_b), and so M increases continuously [179, 180]. The gas thus over-expands to a pressure which is much lower than the background pressure, but yet, at some point in the flow, the flow must adjust to the prevailing background conditions. The resulting dilemma is resolved by the formation of shock waves. These are referred to as 'Mach disk' that is formed normal to the jet centerline, and the 'barrel shock', formed at the sides (Fig. 16.3) [179]. Shock waves reduce the Mach number to subsonic values to meet the prevailing boundary conditions, and once M < 1 the flow can adjust to background conditions, i.e., the isentropic properties of the flow are lost and the pressure and the temperature of the gas in the flow increase.

16.6 Shocks in Supersonic Jets

When an object or a disturbance moves faster than the speed at which information about it can be propagated into the surrounding medium, fluid near the disturbance cannot react and get out of the way before the disturbance arrives thus resulting in a shock wave formation [182]. A shock wave is characterized by an abrupt, discontinuous change in pressure, temperature & even the density of the medium that results in increase in entropy. This can be seen as a phase transition. In order to preserve the cooling produced in the supersonic expansion, the expanding gas should not be allowed to be scattered from the background gas in the expansion chamber. Therefore, the skimmer must be placed upstream of the Mach disk in the continuum region of the flow as shown in Fig. 16.3 for skimming or extracting the beam. The Mach disk location in terms of nozzle diameters is given by the expression, $\frac{x_M}{D} = 0.67\sqrt{\frac{P_0}{P_b}}$ [179], where x_M, the Mach disk location from the nozzle exit and 'D' is the diameter of the nozzle. P_0, P_b, have their usual meanings as discussed earlier. The width (or diameter) of the Mach disk at the front and the barrel shock at the sides are of the order of $0.5x_M$, $0.75x_M$ within an error of ± 25% [179] respectively. Further, the terminal Mach number, M_T, is given by, $M_T = 133\ [P_0D]^{0.4}$, where P_0 is the source or stagnation pressure and D is the diameter of the nozzle as discussed earlier. The variation of Mach number with, x, the distance from the nozzle exit along the jet centerline is given by different relations enumerated in [179]. Once M is known, all other flow parameters in the supersonic jet can be computed.

16.7 Cluster Criteria

The degree of cooling in a free-jet depends on the total number of 2-body collisions that occur during the expansion, which is directly proportional to the product of the stagnation number density and nozzle diameter (P_0D) [183]. Cluster formation requires 3-body collisions and is proportional to P_0^2D. Thus, the ratio of 3-body to 2-body collisions is proportional to stagnation pressure (or number density). By controlling the expanding conditions such that P_0/D is minimum and P_0D is constant, one can obtain in principle, any degree of cooling without any clustering [183]. Except when Helium is used as the carrier gas, clustering effects usually takes place for $P_0D > 10$ torr-cm [172]. Helium, owing to its low boiling point does not undergo clustering easily. Reasonable free-jet continuum properties are obtained for P_0D > 1 torr-cm.

Frequently Asked Questions

Q1. Gas expands isentropically in a nozzle from $P_0 = 3$ bar, $T_0 = 2700$ K. Determine the critical parameters (static temperature, static pressure), the exit velocity and Mach number, at the nozzle exit (assume, specific heat ratio, γ, of the gas to be 1.4).

Q2. Estimate the location of Mach disk for a gas expanding from a nozzle of diameter 150 μm at a stagnation pressure of 3 bar into a chamber maintained at a background pressure of 10^{-2} mbar.

Q3. Calculate the mass flow rate at the sonic exit of a nozzle for a gas mixture of 1% SF_6 in Argon, expanding from a stagnation of 3 bar, 300 K. Diameter of nozzle = 150 μm (specific heat ratio, γ, for SF_6 =1.1 and that for Argon = 1.67, Discharge coefficient = 0.82) [hint: specific heat ratio, γ, of the gas mixture first needs to be evaluated given their mole fractions].