

Chapter 3

Gas Migration Mechanisms

The basic principles and laws governing the migration of natural gases and their seepage to Earth's surface are provided in this chapter by examining the geological factors or processes that influence physical parameters within transport equations. To offer a simple reference framework of seepage processes to readers without specialised knowledge of gas dynamics, migration mechanisms, diffusion and advection in their various forms are summarised without complex mathematics and using carefully controlled terminology. Additional details, retracing the history of gas migration studies, may be obtained from Illing (1933), Muskat (1946), MacElvain (1969), Bear (1972), Pandey et al. (1974), Malmqvist and Kristiansson (1985), Price (1986), and Brown (2000), as well as in the review paper by Etiope and Martinelli (2002). Oil migration is not the focus of this chapter.

3.1 Fundamentals

3.1.1 Sources and Pathways

Gas migration and seepage to the surface are strictly related to the existence of two geological features, a gas migration source and a preferential route for gas motion. The concepts are linked to the Petroleum Seepage System (PSS) introduced in Chap. 1.

The migration source (the starting point of migration) is not necessarily the gas source, in other words, where the gas is generated (source rock). Accumulations of gas (reservoirs) are common migration sources. However, it is possible that gas seeping to Earth's surface may also come directly from source rocks (e.g., shales) without reservoir intermediation, as considered in recent theoretical models (Berbesi et al. 2014) and suspected in a seep located in the State of New York, USA (Etiope et al. 2013). As discussed in Chap. 5, this argument is fundamental for assessing petroleum systems.

The preferential routes of gas flow are zones of enhanced permeability such as sand horizons within a clayey sequence (mainly horizontal migration), and tectonic discontinuities such as faults and fracture networks (mainly vertical migration). Permeability is the basic parameter controlling gas flow through porous and fractured media (porosity only determines the gas volume stored within a rock). Permeability is a constant determined only by the media's structure and independent of the nature of the fluid passing through the media (Muskat 1946). Hence, water permeability and gas permeability are the same for dry media. For a two-phase system, gas permeability decreases as water content increases because the space available for gas is reduced.

In practice, secondary permeability due to the fracturing and faulting induced by tectonic movements is the leading factor driving gas seepage. The shape, size, and local distribution of macro-seeps, as well as gas injection tests (e.g., Ciotoli et al. 2005) and “chimneys” in seismic images (e.g., Heggland 1998; Loseth et al. 2009), indicate that gas typically follows channels of enhanced permeability (i.e. of minor resistance to gas motion), and that once the preferential pathway (the ‘channel’) is activated, gas flow only occurs along this channel and is insensitive to the permeability of contiguous rock volumes. The process is also known as “fracture flow” (Loseth et al. 2009). The channel provides minimum dissipation for gas energy and the conservation of pressure and flow rate. Basically, seepage does not occur throughout the fault line but only in some portions, forming spotty seeps on the surface. Channels may then “migrate” horizontally and change their position along faults due to, for example, fracture self-sealing and fracture propagation. In fact, it is not rare to observe seeps (vents and craters) that change their location by a few meters or tens of meters year after year.

3.1.2 Diffusion and Advection

Depending on the source and the surrounding permeabilities, gas movement can be induced by two types of force fields, concentration gradients and pressure gradients. In the first case, the spreading of gas molecules in a direction that tends to equalise the concentrations in all parts of a rock system occurs as gas “diffusion”. In the second case, the entire gaseous mass tends to move from a zone of high pressure to a zone of low pressure; this mass transport is called “advection”.

In the scientific literature, the terms “mass transport”, “viscous flow”, “fluid flow”, “air flow”, “non-diffusive transport”, and “effusion” (e.g., Harbert et al. 2006) are also used for advection. However, some authors have improperly used the term “convection” to indicate pressure-driven transport (e.g., Mogro-Campero and Fleischer 1977). “Convection” is advective movement with a pressure gradient generated by thermal gradients. Since it disperses more rapidly and consequently become lighter, a warmer gas ascends (i.e., at constant volume, a warmer gas is more pressurised). In other words, “convection” is a form of advection driven by temperature gradients. Changing the perspective from temperature to pressure is

possible using the equation of state. It is incorrect, however, to name as “convective” something that is not linked to temperature effects, such as normal gas flows linked to buoyancy or to hydrostatic and lithostatic gradients. Convective flows are more typical of geothermal systems.

Diffusion and advection can be examined using transport equations without complex mathematics, i.e. assuming realistic limitations (those frequently adopted to solve practical problems) on the nature of fluid and porous media. In Muskat (1946), Wickoff outlines the inadequacy of applying the rigorous mathematical solutions of certain physical laws to complex geological settings.

Diffusion is the movement of a chemical species from a volume of high concentration of that species to a volume of lower concentration. The movement is described by Fick’s Law, in which the gas flux is directly related to the concentration gradient and a constant, as follows:

$$F = -D_m \nabla C \quad (\nabla = \delta/\delta x + \delta/\delta y + \delta/\delta z) \quad (3.1)$$

or for a one-dimensional form along the z axis, as follows:

$$F = -D_m dC/dz \quad (3.2)$$

where D_m is the molecular diffusion coefficient (m^2/s) and dC is the variation in gas concentration (kg/m^3) along dz (m). The molecular diffusion coefficient is a constant for the specific gas and it is controlled by molecular size and shape. The lighter the hydrocarbon, the greater the coefficient. The coefficient only changes with temperature, pressure, and the physical nature of the substance through which molecular motion occurs. In rock pores, this substance is generally water or air (or a gas mixture). Therefore, for each gas, the diffusion coefficient in water (D_{mw} or simply D_w) must be distinguished from the diffusion coefficient in air (D_{ma} or simply D_m). Furthermore, in porous media the volume through which gas diffuses is reduced and the average path length between two points is increased (i.e. tortuosity). Interstitial diffusion is then defined by the “effective” diffusion coefficient (D_e), as follows:

$$D_e = D_m n \quad (3.3)$$

where n is the effective porosity of the media (%) and describes diffusion by considering the motion of a gas molecule through a porous structure. Global diffusion is defined by the “apparent” diffusion coefficient (D), also known in the literature as the “true” or “bulk” coefficient; it includes the effects of the porosity and tortuosity of the media. For soil, most authors agree to define this coefficient, as follows (e.g., Lerman 1979):

$$D = D_e n = D_m n^2 = D_m n/\tau \quad (3.4)$$

where τ is the tortuosity of the media. In conclusion, $D_m > D_e > D$.

In practice, a gas diffusing over time t will cover a diffusive distance (Z_d) based on the following equation:

$$Z_d = (D t)^{0.5} \quad (3.5)$$

The equation indicates that methane diffusing in still water (D_w : 1.5×10^{-5} cm²/s) will cover 1.1 cm in 1 day, 22 cm per year, 6.9 m in 1000 years, etc. Ethane, propane, and butane will be progressively slower, as the diffusion coefficient of each hydrocarbon is 1.23 times that of its next heavier homolog (e.g., Witherspoon and Saraf 1965).

The term advection refers to the movement of matter under the influence of external forces, namely pressure gradients. In a broader sense, all of the following movements due to “global” forces are advective (Lerman 1979): atmospheric precipitation, evaporation, wind, the deposition of sediments, groundwater flow, and the movement of crustal plates.

A gas with a concentration C (kg/m³) and a velocity v (m/s) results in the following flux:

$$F = C v \quad (3.6)$$

Velocity depends on the pressure gradient and on a mobility coefficient that is related to the geometry of the media and gas viscosity. In the case of advection through dry porous media, the mobility coefficient depends on the intrinsic permeability of the media, according to Darcy’s Law:

$$v = -k\nabla P/\mu \quad (\nabla = \delta/\delta x + \delta/\delta y + \delta/\delta z) \quad (3.7)$$

or in a one-dimensional form, along the z axis, and for a short distance:

$$v = k\Delta P/(\mu Z) \quad (3.8)$$

where v is the gas velocity (m/s), k is the intrinsic permeability (m²), μ is the dynamic gas viscosity (kg m⁻¹ s⁻¹), and ΔP is the pressure difference (kg m⁻¹ s⁻²) between two points spaced at a distance Z (m).

An estimate of advective gas velocity through a planar fissure may be obtained using the following formula (Gascoyne and Wuschke 1992):

$$v = (b^2/12\mu)(dP/dz) \quad (3.9)$$

where $(b^2/12)$ is the fissure permeability, b is the fissure width, and μ is the gas viscosity.

For estimating gas velocity through fractured media (a system of intersecting fissures), “cubic law” (Schrauf and Evans 1986) can apply, as follows:

$$v = (b^3/6d\mu)(dP/dz) \quad (3.10)$$

where d is the mean distance between intersecting fissures (m).

Advective processes can occur within the subsurface whenever pressure gradients between two points occur. Such gradients can be induced by tectonic stresses, variations in lithostatic loading, rock fracturing, localised gas generation, the recharge and discharge of aquifers and deep fluid reservoirs, and, near the surface, by barometric pressure pumping. Also, the natural tendency of lighter gases (e.g., helium, hydrogen) to ascend due to low density is an advective phenomenon. In fact, a gas with a density ρ_1 , moves upward if it is bounded by a gaseous phase with a density $\rho_2 > \rho_1$. The lighter gas is subjected to a pressure gradient $\rho_2 g$ and the following equation is valid:

$$v = k g(\rho_2 - \rho_1)/\mu \quad (3.11)$$

with g acceleration of gravity, and the term $g(\rho_2 - \rho_1)$ is equivalent to a pressure gradient. The very existence of a naturally occurring background pressure gradient in the Earth is an index of continuous outgassing.

In the geological environment, diffusion and advection almost never act separately; thus, formally, the movement of gas should be ascribed to them in combination. By combining the diffusion and advection terms, the total flux of gas, in a one-dimensional form, is:

$$F = [-n D_m(dC/dz)] + [v C] \quad (3.12)$$

where $[-n D_m(dC/dz)]$ is the diffusive term and $[v C]$ is the advective term.

Depending on the assumptions and limitations adopted, the general equation of transport, in terms of mass conservation, may be written in more or less complex forms. In most cases, for practical problems, migration models and their relevant equations can follow criteria of simplicity and acceptable approximation. Therefore, it is possible to consider one-dimensional equations for laminar and steady-state flows through dry, homogeneous, and isotropic porous media.

Hence, the following general transport equation, common in fluid mechanics, can be obtained:

$$n D_m(d^2C/dz^2) - v(dC/dz) + (\alpha - \omega) = 0 \quad (3.13)$$

where α is the generation rate of the gas and ω is the rate of removal of the gas from the stream (as a result of adsorption by rocks, the dissolution by groundwater, or microbiological consumption). In practice, several variants of the equation exist.

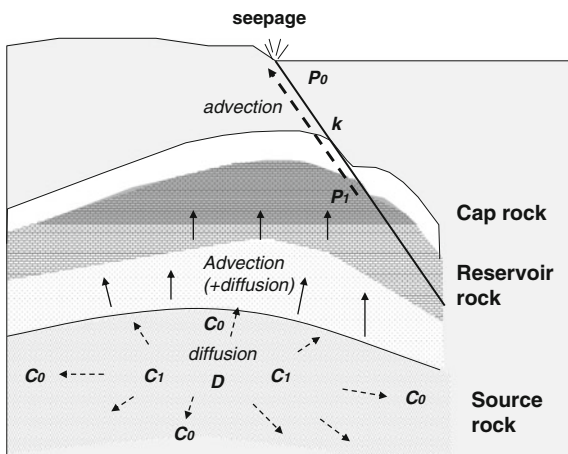
3.2 Actual Mechanisms and Migration Forms

For a seepage system especially, the velocity and spatial scales of advective movements are much higher than the diffusive ones. Diffusion is only important in capillaries or small-pore rocks, indicating that within a petroleum seepage system diffusion is the prevailing mechanism of “primary migration” (Hunt 1996) in gas source rocks (shales and limestones), after or during the generation of hydrocarbons. Thus, the diffusion coefficients of methane and heavier alkanes in water (mentioned above) should be considered. Frequently, the generation of hydrocarbons is concurrent with the diagenesis of clays such as smectites to illite, resulting in water being simultaneously driven out as an advective process. Some gas can then be transported in solution by water. The passage of gas from source to external rocks to trapped reservoir rocks (secondary migration), and from one reservoir to another or to Earth’s surface (tertiary migration) is largely driven by pressure gradients (Fig. 3.1). Advection assumes an exclusive role in larger pores or in fractured media outside of source rocks.

However, depending on the physical-geological conditions that the gas encounters, the nature of the driving force can change during gas ascent. Furthermore, sedimentary basins and their constituent rocks are subjected to basin loading, compaction with water loss, extensional and compressional stresses, and other tectonic forces that change the driving force of gas flow. Finally, variations in temperature, pressure, mechanical stress, chemical reactions, and mineral precipitation change the gas-bearing properties of geological formations. Interactions between all of these factors may lead to time-dependent fluid transport for which gas seepage may be quite variable on daily, seasonal, and geological time scales.

Depending on gas-water-rock system conditions, gas advection can have different forms (Fig. 3.2). In dry porous or fractured media, gas flows through interstitial or fissure space and is defined as gas-phase advection. Equation (3.7) can

Fig. 3.1 The main factors controlling gas migration from source rocks to reservoirs and to the surface (P gas pressure; k permeability; D gas diffusion coefficient; and C gas concentration)



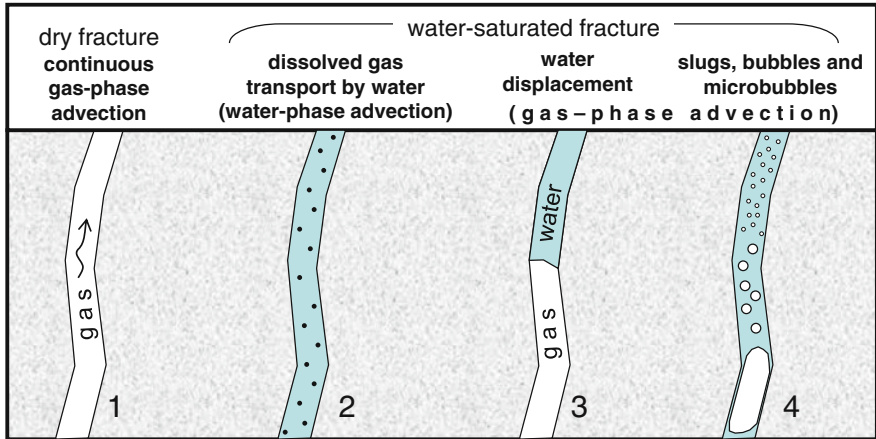


Fig. 3.2 A schematic picture of gas advection forms in dry and water-saturated fractured media. 1 Pressure-driven continuous gas-phase flow through dry fractures. 2 The water-phase transport of gas in solution. 3 A pressure-driven or density-driven continuous gas-phase displacing water in saturated fractures. 4 The buoyant movement of gas bubbles in aquifers and water-filled fractures, either as slugs or microbubbles

be applied. For saturated porous media, three possible phenomena may occur, as follows:

- (a) Gas dissolves and is transported by groundwater (water-phase advection);
- (b) Gas flows and displaces water (gas-phase advection); and
- (c) Gas flows as bubbles.

In water-phase advection (a), the gas, being in solution, moves at the same velocity as water; hence, Darcy’s equation, in the following form used in hydrogeology, is valid:

$$v = K i \tag{3.14}$$

where K is the hydraulic conductivity of the media (in darcy units) and i is the hydraulic gradient.

For gas-phase advection through water-saturated media, gas must have a pressure (P_g) above the sum of hydrostatic pressure (P_w) plus a capillary pressure (P_c). Hydrostatic pressure is given by the height of the piezometric surface (H_w) from the point considered ($P_w = \rho_w g H_w$). Capillary pressure is linked to the interfacial tension of water (σ) and to the pore throat radius (r) according to the Laplace equation ($P_c = 2\sigma/r$). When $P_g < (P_w + P_c)$, gas only enters the media by diffusion. When $P_g > (P_w + P_c)$ two-phase flow occurs, with water displaced by gas. If $P_g = P_{fr} \gg (P_w + P_c)$ gas fractures the rock (P_{fr} is the pressure at which the fracture begins and roughly corresponds to the lithostatic pressure). When the gas pressure P_g reaches P_{fr} , gas flows through the fracture planes generated. If, however, $P_{fr} < (P_w + P_c)$, gas flow only occurs in the fracture and there is no

migration of gas within the rock matrix. The pressure can rise and bring about the propagation of the fracture network. If, on the contrary, $P_{fr} > (P_w + P_c)$, gas will flow in the fracture and from the fracture towards the matrix. Here, it should be noted that both hydrostatic and lithostatic pressure can operate on the gas (as occurs in a “gas cap”) to serve as a driving force for the gas itself.

As a result, water displacement (b) occurs when $P_g > (P_w + P_c)$ and can be at a different scale depending on the dimension of the advancing front of the gas with respect to the type of water-bearing media (homogenous porous media, single fracture, etc.). For example, within a saturated fissure, gas totally displaces water if the gas strip front has a size similar to the fissure width (Gascoyne and Wuschke 1992). Equation (3.9) can be used by considering the difference of density between gas and water as the pressure gradient. On the contrary, if gas moves as a tiny strip, with a size less than that of the fracture width, or moves as an intermittent flow (i.e. P_g varies in time from values above to values below the displacement threshold $= P_w + P_c$) or, finally, exsolves from water by oversaturation, then gas bubbles form (c). The bubble flow mechanism is considered to be a common form of gas movement in the subsurface, so deserves a separate discussion.

3.2.1 Bubble and Microbubble Flow

Bubble and microbubble buoyancy is a commonly proposed mechanism for gas seepage, including microseepage (Price 1986; Klusman and Saeed 1996; Saunders et al. 1999; Brown 2000). The phenomenon has been studied in its two main stages, bubble formation and bubble motion.

Bubbles in a liquid can originate from two main mechanisms: (a) the supersaturation of a solution of a gas or (b) direct introduction into a liquid (i.e. mechanical entrainment; Teller et al. 1963). Saturation depends on gas solubility, which is, in turn, influenced by temperature and pressure (as well as by pH and ionic strength). Temperature increases and pressure decreases lead to water degassing. In the subsurface, depressurisation can occur when moving groundwater meets fractures, when the hydrostatic or lithostatic pressure drops (e.g., after erosion, landslides, or mining), or when pumping effects are induced by tectonic stress or aquifer charging and discharging. Increasing temperatures can be linked to variations in the thermal conductivity of rocks (i.e., water crosses rocks with different thermal conductivities) or to variations within the local geothermal gradient resulting from hydrogeological or tectonic phenomena. The appearance of a bubble requires a gas nucleus as a void in the liquid. The nucleus may be in the form of a nanoscopic bubble or of a solid carrying adsorbed gas. Such “heterogeneities” act as catalysts for the degassing induced by T and P variations. For example, bubble formation is made easier by energetic particles produced during the “alpha” decay of radioactive minerals (Malmqvist and Kristiansson 1985). Obviously, a solution also becomes oversaturated when gas is locally generated by microbiological or

chemical reactions. For further details on bubble nucleation and formation, the reader may refer to Frenkel (1955), Mesler (1986), and Tsuge (1986).

Bubbles and microbubble streams can form when a fault crosses an aquifer, with gas coming from depth along the fault itself. The growth of a gas bubble in water is controlled by the diffusion of the gas dissolved in water, and by the decompression of gas within the bubble as the hydrostatic pressure decreases. With bubble growth, buoyancy forces become more important and at a certain moment overcome the drag forces that tend to keep the bubble stationary. A bubble in water can then move in accordance with Stokes' Law, as follows:

$$v = d^2 g (\rho_w - \rho_g) / 18 \mu_w, \quad (3.15)$$

where v is the bubble velocity (m s^{-1}), d is the bubble diameter (m), g is the gravity acceleration (m s^{-2}), ρ_w and ρ_g are the water and gas density, respectively (kg m^{-3}), and μ_w is the water viscosity ($\text{kg m}^{-1} \text{s}^{-1}$). The equation indicates that the velocity of a bubble is directly related to the square of its diameter. When hydrostatic pressure decreases, d increases and bubbles accelerate with respect to the surrounding water. The equation, so written, is the general form of Stokes' Law. The equation must be properly modified for porous media. First, parameter d must have an upper limit somehow related to the structure of the media. Explicitly, it is expected that the maximum size of bubbles is controlled by the minimum transverse section of the migration path through porous media. For a fractured rock, bubble size may be related to the minimum distance between the fissure walls. Using the following equation, Várhegyi et al. (1986) described a theoretical model for estimating the size of a bubble (d_B) and its velocity as a function of media porosity (n) and grain diameter (d_G):

$$d_B = 1.26 d_G n(n + 0.21) \quad (3.16)$$

Using this formula, it is possible to derive the maximum velocity (with the bubble size being equal to the pore space) of gas bubbles through homogeneous and equigranular porous media, although this kind of media is only rarely found in nature. In theory, the relationship between d_G and the true grain size distribution is very difficult to investigate. However, for the case of a wide grain size distribution, it is likely that d_G (equivalent to mean grain size) is shifted towards finer sizes and that the cross section available for bubble flow is reduced (Várhegyi et al. 1986). To estimate the order of magnitude of microbubble velocity in geologic media, the modified Stokes' equation (3.16) may be used, although the model was developed by considering the generic Stokes' Law of bubble motion using bubble diameter as a function of rock porosity. For fractured media, the fracture or fissure width determines the maximum bubble diameter required for Várhegyi's equation. Since it does not take into account a number of factors that occur under real conditions, the simple model must be considered as a first approach for bubble velocity derivations in geological environments. The velocity given by Stokes' equation should refer to single bubbles in "unbounded" water conditions, when the motion and

shape of a given bubble is not perturbed by other bubbles or the wall effect induced by the fracture. By increasing gas fluxes, bubbles can coalesce to produce vertically elongated bubbles called “slugs”, then continuous gas streams within the fracture.

In short, depending on the gas flux and fracture size for which the velocity of gas bubbles must be examined, the following bubble flow patterns can be identified as those possibly occurring in natural rock fractures:

1. Bubbles with a negligible fracture wall effect. Classic equations of single bubble motion can be used by assuming that there is no perturbation of bubble flow by the fracture walls. Such a condition can occur for microbubbles in relatively larger fractures and rock voids.
2. Bubbles rising along a typically narrow fracture whose walls influence bubble rise (the fracture width close to the bubble diameter). The bubble velocity (v_w), normalized to Stokes' velocity (v), depends on the ratio of the bubble's radius (r) to the half width (b) of the fracture (approximated by parallel plates) according to Brown (2000), as follows:

$$\frac{v_w}{v} = 1 - 1.004\left(\frac{r}{b}\right) + 0.418\left(\frac{r}{b}\right)^3 - 0.21\left(\frac{r}{b}\right)^4 - 0.169\left(\frac{r}{b}\right)^5 \quad (3.17)$$

3. Long bubble-trains and slugs ($r > b$). By increasing gas flux and/or reducing the fracture aperture, bubbles become elongated (slugs) forming a typical bubble-train flow.
4. Bubble plumes in larger rock voids ($r \ll b$). Due to bubble turbulence, an additional upwelling fluid velocity should be considered (variable from 10 to 40 cm/s) (Clift et al. 1978). In large joint systems, water-filled cavernous zones, and sinkholes in karst environments, intense bubble plumes can rise without significant wall friction.

At higher gas pressures and fluxes, slugs can be replaced by connected gas streams driven by pressure gradients. In particular, bubbles coalescing within a subsequent gas stream can occur if the pressure-driven velocity is higher than the buoyancy-driven velocity. Bubble-trains and slug flow can be due to intermittent gas leakages through reservoir–cap rock systems or to the transmission of pressure pulses created by crack propagation due to tectonic (seismic) stresses. Continuous phase flow may exist only if the fracture is continuously invaded by large amounts of gas, with pressures above hydrostatic plus capillary pressures (e.g., leakage from geothermal or hydrocarbon pressurized reservoirs). Any reduction of gas pressure or fracture width will interrupt the flow, and slugs or trains of bubbles will form. As a bubble rises, its radius increases and can be occluded within the fracture. As bubbles occlude, they again coalesce to form longer slugs and then continuous phase gas columns.

3.2.2 Gas Seepage Velocity

Obtaining the velocity of gas migration is an important task in gas seepage studies. Theoretical gas velocity as a function of fracture width can be calculated using Eq. (3.9) for continuous gas-phase flow between parallel plates and Eq. (3.15) for bubble flow in the Stokes regime, by assuming that the bubble diameter is smaller than the fracture width. In Fig. 3.3, velocity curves were plotted for reference conditions corresponding to a subsurface depth of 1,000 m (i.e., 38 °C and 10 MPa; a water density of 1,000 kg/m³; a water viscosity of 0.0009 Pa; a gas density of 100 kg/m³; and a gas viscosity of 0.000015 Pa). Due to the simple buoyancy of gas in water, the pressure gradient is assumed to be density-driven. Bubble velocity was computed both with and without the wall effect (Eqs. 3.15 and 3.17, respectively).

Experimental data on in situ gas velocity are very difficult to obtain. Very few examples that include fracture data are available within the literature. Those that are available come primarily from studies on the sealing properties of gas storage reservoirs (e.g., Jones and Thune 1982), or from field gas injection tests (generally performed as a part of studies on the geological disposal of radioactive wastes; e.g., Lineham et al. 1996; Ciotoli et al. 2005) where the injected gas has a pressure equal to the hydrostatic pressure plus a capillary pressure. Studies of leakage from gas storage reservoirs and controlled experiments suggest gas velocities of hundreds of

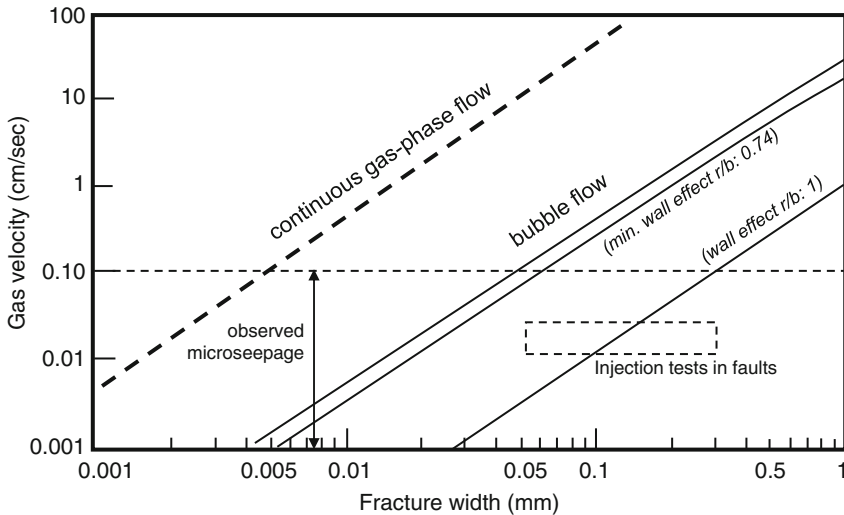


Fig. 3.3 Plot of gas velocity as a function of fracture width (redrawn from Etiope and Martinelli 2002). The theoretical velocities of continuous gas-phase flow and bubble flow were computed for fluid properties at a depth of 1,000 m (see text). The wall-effect bubble velocity was computed for $r/b = 0.74$ and $r/b = 1$. The rectangle represents experimental values from a gas injection test along a fault (Ciotoli et al. 2005). The range of observed microseepage velocities was obtained from Brown (2000)

centimeters per day, orders of magnitude greater than those driven by diffusion alone (Jones and Thune 1982; Harbert et al. 2006). Some conservative estimates for velocity, using an unknown fracture aperture, were made by evaluating the effects of subsurface pressure changes on the surface geochemical signatures of hydrocarbon seepage (Brown 2000). In special cases related to gas vents, velocities can be estimated by measuring the flux of the gas emitted; 150–300 m/day was conservatively estimated for gas rising through mud volcanoes (Martinelli and Ferrari 1991).

Theoretically, as demonstrated by Brown (2000), continuous phase gas migration is the fastest mechanism. In fact, the velocity of continuous phase flow is controlled by the viscosity of gas (Eq. 3.7). The viscosity controlling bubble ascent is that of water, which is approximately 60 times that of gas using the assumed reference conditions. For fractures of a few millimetres, bubble velocity ranges from 0.001 to 10–20 cm/s. Microbubbles of colloidal size (radii below 1 μm), considered by MacElvain (1969) and Price (1986) to be a favourable mechanism for hydrocarbon gas transport, should have low velocities on the order of 10^{-6} – 10^{-5} cm/s. Instead, observed gas velocities range on the order of 10^{-4} – 10^0 cm/s (0.1–2,000 m/day). Figure 3.3 suggests that these velocities can easily be reached by continuous phase flows at any fracture width and by bubbles within fractures larger than 0.01 mm. For larger fracture apertures and voids on the order of cm, microbubble plumes may reach velocities on the order of 10^4 m/day. Depending on the wall effect, bubble-trains and slugs can have velocities intermediate between microbubbles and continuous gas flow. Heinicke and Koch (2000) observed that hydrogeochemical earthquake signals can be due to CO_2 slugs rising through water-filled faults at velocities of approximately 7–8 cm/s (6,000–7,000 m/day). The conclusion of Brown (2000), that gas-bubble ascent cannot account for observed microseepage velocities, is, therefore, only valid for bubbles of colloidal size.

Overall, field and laboratory gas injection tests (e.g., Etiope and Lombardi 1996; Ciotoli et al. 2005) indicate that aquifers do not constitute a barrier for gas migration or reduce gas velocity. In fact, due to the higher buoyancy that develops between gas and water (Eq. 3.15) as compared to that between gas and gas (Eq. 3.11), under the same injection pressure gas in saturated rock moves faster than gas in dry rock.

3.2.3 Matter Transport by Microbubbles

It was suggested, even if experimental data are still scarce, that during their ascent in fractured rocks microbubbles can transport gaseous and solid material, including metallic minerals and radionuclides. This process is generally described as “matter transport by geogas” (Malmqvist and Kristiansson 1984; Kristiansson et al. 1990; Hermansson et al. 1991; Etiope 1998). Since the particles may include precious minerals, such as gold, copper and zinc, and radioactive elements, such as uranium, radium and cesium, this phenomenon may have several implications both in ore

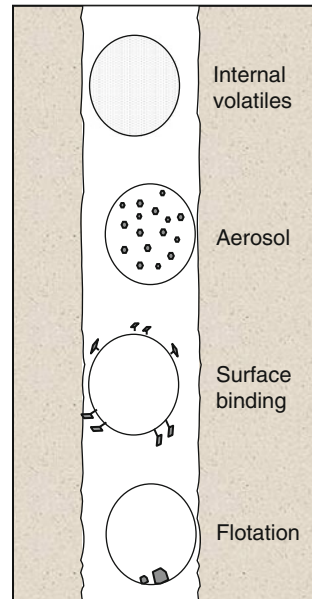
prospecting and in underground nuclear or toxic waste repositories. Bubble can take (strip) gaseous and solid material from the surrounding rocks and transport them through four types of physical mechanisms (Fig. 3.4; Hermansson et al. 1991; Etiope 1998): (a) flotation (lifting of solid particles inside the bubble); (b) binding on the gas–water interface of active elements; (c) aerosol transport; (d) transport of volatile compounds dissolved in the carrier gas.

Flotation is a well known physical process (e.g., Aplan 1966) due to the fact that the specific surface energy is higher between water and gas than between minerals and gas. Thus a microbubble stream crossing crushed rocks can lift fine particles and transport them upward. Laboratory observations effectively proved that gas bubbles in porous media can transport argillaceous particles (Goldenberg et al. 1989), and fine powder of metallic elements and radionuclides (Etiope and Lombardi 1996).

Transport of active elements on gas–water interface is due to the lower energy level provided by the interface itself than that occurring in solution. Many elements, mainly radionuclides, tend to attach and concentrate on bubble surfaces (Peirson et al. 1974; Pattenden et al. 1981). It has been shown, for example, that a substantial enrichment of elements can take place on surfaces of bubbles of air that pass through seawater. It was found that foam on the surface water contains up to 600 times more plutonium per unit volume than the sea water (Walker et al. 1986).

Aerosol transport may occur by dispersion of solid and/or liquid particles induced by rapid movement of gas pockets through the rocks. This mechanism is also known as “geoaerosol transport” (Holub et al. 2001) and aerosol sampling from

Fig. 3.4 The four mechanisms of matter transport by gas microbubbles along rock fractures



rock fissures has been used in mineral exploration (Kristiansson et al. 1990; Kremár and Vylita 2001).

Finally, volatile compounds can be mixed inside the bubble gas. They may include both gaseous molecules and volatile compounds such as mercury and arsenic. If such compounds are formed in the fractures of the rocks, they may dissolve in the geogas and be transported to the surface.

These bubble transport mechanisms, especially flotation, appear to be also important for the rapid migration of liquid hydrocarbons and oil. In many mud volcanoes it is not rare to observe oil droplets and iridescences around bubble plumes (Fig. 3.5). Specific studies are however necessary to better understand and model the phenomenon.

3.2.4 The Concept of Carrier Gas and Trace Gas

Another important physical concept regarding gas migration is that of the transport of trace gases by a carrier gas. Advective migration discussed above, either as continuous gas flow or microbubbles, requires a stream of “free gas”, i.e. gravitative forces act only on gases which occur at sufficiently large concentrations (gas domain). To form a stream of a particular gaseous species, an immense number of molecules of that species must be available at the same location at the same time. In many cases the amount of heavy gaseous alkanes (especially propane, butane and pentane) and noble gases, such as helium (He) and radon (Rn), occurring in the subsurface is many orders of magnitude too small (orders of ppmv or 10^{-10} ppm for Rn) to form a macroscopic quantity of gas which can react to pressure gradients and flow autonomously by advection (pure radon or helium bubbles do not exist!). Such gases must be carried by a macroscopic flow of another gas which is moving upwards, i.e. a “carrier gas”, able to form large gaseous domains. Carrier gas is generally CH_4 , CO_2 or N_2 , depending on the specific geological environment. Anomalous concentrations of helium in soil-gas (discussed in Chap. 5), for example, can only be explained by an ascending carrier gas, and in fact helium in soil or groundwater is always associated to a major gas, such as CH_4 , N_2 or CO_2 . The long-distance transport of radon is another phenomenon due to carrier gas advection (e.g., Malmqvist and Kristiansson 1984; Etiope and Martinelli 2002). Radon (^{222}Rn) is an unstable nuclide with a short half-life (3.8 days), so its concentration rapidly decreases during the slow diffusion in the rocks. In order for radon to reach the surface from deep sources (uranium or radium bearing rocks or fluids) before decaying, it must be transported upwards at a rapid rate, which, in itself, is possible only if a rapidly ascending carrier gas exists.

This concept is quite important when applied to the evaluation of sources of the several hydrocarbons occurring in a seeping gas. The minor hydrocarbon compounds that may occur at trace levels (ppmv or ppbv) in a surface seep, such as butane, pentane or olefins (i.e., alkenes, such as ethene, propene, butene) cannot move rapidly and for long distances autonomously. In a gaseous flow system



Fig. 3.5 Oil layers and iridescences around bubbles in mud volcano craters (**a** Kechaldag, Azerbaijan; *photo* by L. Innocenzi, INGV; **b** Paclele Mici, Berca, Romania; *photo* by L. Spulber)

methane is always their carrier gas. This implies that their origin (source rock) cannot be very distant from that of methane, and in most case it is the same. Such a type of evaluation is important to assess the origin of the gas, the possible mixing of compounds from different sources and, thus, the petroleum system, as discussed in Chap. 5.

References

- Aplan FF (1966) Flotation. In: Kirk-Othmer (ed) Encyclopedia of chemical technology, 2nd edn. Wiley-Interscience Publications, New York
- Bear J (1972) Dynamics of fluids in porous media. Elsevier, New York
- Berbesi LA, di Primio R, Anka Z, Horsfield B, Wilkes H (2014) Methane leakage from evolving petroleum systems: masses, rates and inferences for climate feedback. *Earth Planet Sci Lett* 387:219–228
- Brown A (2000) Evaluation of possible gas microseepage mechanisms. *AAPG Bull* 84:1775–1789
- Ciotoli G, Etiopie G, Guerra M, Lombardi S, Duddridge GA, Grainger P (2005) Migration and behaviour of gas injected into a fault in low-permeability ground. *Q J Eng Geol Hydroge* 38:305–320
- Clift R, Grace JR, Weber ME (1978) Bubbles, drops and particles. Academic Press, New York
- Etiopie G (1998) Transport of radioactive and toxic matter by gas microbubbles in the ground. *J Environ Radioact* 40:11–13
- Etiopie G, Lombardi S (1996) Laboratory simulation of geogas microbubble flow. *Environ Geol* 27:226–232
- Etiopie G, Martinelli G (2002) Migration of carrier and trace gases in the geosphere: an overview. *Phys Earth Planet In* 129:185–204
- Etiopie G, Drobniak A, Schimmelmann A (2013) Natural seepage of shale gas and the origin of “eternal flames” in the Northern Appalachian Basin, USA. *Mar Pet Geol* 43:178–186
- Frenkel J (1955) Kinetic theory of liquids. Dover, New York
- Gascoyne M, Wuschke DM (1992) Gas flow in saturated fractured rock: results of a field test and comparison with model predictions. In: “Gas generation and release from Rad.Waste Rep.”, Proceedings of the NEA Workshop, Aix en Provence, 23–26 Sept 1991
- Goldenberg LC, Hutcheon I, Wardlaw N (1989) Experiments on transport of hydrophobic particles and gas bubbles in porous media. *Transport Porous Med* 4:129–145
- Harbert W, Jones VT, Izzo J, Anderson TH (2006) Analysis of light hydrocarbons in soil gases, Lost River region, West Virginia: relation to stratigraphy and geological structures. *AAPG Bull* 90:715–734
- Heggland R (1998) Gas seepage as an indicator of deeper prospective reservoirs. A study on exploration 3D seismic data. *Mar Pet Geol* 15:1–9
- Heinicke J, Koch U (2000) Slug flow—a possible explanation for hydrogeochemical earthquake precursors at Bad Brambach, Germany. *Pure Appl Geophys* 157:1621–1641
- Hermansson HP, Sjoblom R, Akerblom G (1991) Geogas in crystalline bedrock. Statens Kärnbränsle Nämnd (SKN) report 52, Stockholm
- Holub RF, Hovorka J, Reimer GM, Honeyman BD, Hopke PK, Smrz PK (2001) Further investigations of the ‘Geoerosol’ phenomenon. *J Aerosol Sci* 32:61–70
- Hunt JM (1996) Petroleum geochemistry and geology. W.H. Freeman and Co, New York, 743 pp
- Illing VC (1933) Migration of oil and natural gas. *Inst Petrol Technol J* 19:229–274
- Jones VT, Thune HW (1982) Surface detection of retort gases from an underground coal gasification reactor in steeply dipping beds near Rawlins, Wyoming. In: Society of Petroleum Engineers, SPE paper 11050, p 24

- Klusman RW, Saeed MA (1996) A comparison of light hydrocarbon microseepage mechanisms. In: Schumacher D, Abrams MA (eds) Hydrocarbon migration and its near surface effects. AAPG Memoir 66:157–168
- Krcmár B, Vylita T (2001) Unfilterable “geoaerosols”, their use in the search for thermal, mineral and mineralized waters, and their possible influence on the origin of certain types of mineral waters. *Environ Geol* 40:678–682
- Kristiansson K, Malmqvist L, Persson W (1990) Geogas prospecting: a new tool in the search for concealed mineralizations. *Endeavour* 14:28–33
- Lerman A (1979) *Geochemical Processes, water and sediment environments*. Wiley Interscience Publications, New York
- Lineham TR, Nash PJ, Rodwell WR, Bolt J, Watkins VMB, Grainger P, Heath MJ, Merefield JR (1996) Gas migration in fractured rock: results and modelling of a helium gas injection experiment at the Reskajeage Farm Test Site, SW England, UK. *J Contam Hydrol* 21:101–113
- Losef H, Gading M, Wensaas L (2009) Hydrocarbon leakage interpreted on seismic data. *Mar Pet Geol* 26:1304–1319
- MacElvain R (1969) Mechanics of gaseous ascension through a sedimentary column. In: Heroy WB (ed) *Unconventional methods in exploration for petroleum and natural gas*. Southern Methodist University Press, Dallas, pp 15–28
- Malmqvist L, Kristiansson K (1984) Experimental evidence for an ascending microflow of geogas in the ground. *Earth Planet Sci Lett* 70:407–416
- Malmqvist L, Kristiansson K (1985) A physical mechanism for the release of free gases in the lithosphere. *Geoexploration* 23:447–453
- Martinelli G, Ferrari G (1991) Earthquake forerunners in a selected area of northern Italy: recent developments in automatic geochemical monitoring. *Tectonophysics* 193:397–410
- Mesler R (1986) Bubble nucleation. In: Cheremisinoff NP (ed) *Gas-liquid flows*, Encyclopedia of fluid mechanics, vol 3, Gulf Pub Co
- Mogro-Campero A, Fleischer RL (1977) Subterrestrial fluid convection: a hypothesis for long-distance migration of radon within the Earth. *Earth Planet Sci Lett* 34:321–325
- Muskat M (1946) *The flow of homogeneous fluids through porous media*. Edwards Inc., Ann Arbor, Michigan
- Pandey GN, Rasintek M, Katz DL (1974) Diffusion of fluids through porous media with implication in petroleum geology. *AAPG Bull* 58:291–303
- Pattenden NJ, Cambay RS, Playford K (1981) Trace elements in the sea-surface microlayer. *Geochim Cosmochim Acta* 45:93–100
- Peirson DH, Cawse PA, Cambay RS (1974) Chemical uniformity of airborne particulate material and a maritime effect. *Nature* 251:675–679
- Price LC (1986) A critical overview and proposed working model of surface geochemical exploration. In: *Unconventional methods in exploration for petroleum and natural gas IV*, Southern Methodist University Press, pp 245–309
- Saunders D, Burson KR, Thompson CK (1999) Model for hydrocarbon microseepage and related near-surface alterations. *AAPG Bull* 83:170–185
- Schrauf TW, Evans DD (1986) Laboratory studies of gas flow through a single natural fracture. *Water Resour Res* 22:1038–1050
- Teller AJ, Miller SA, Scheibel EG (1963) Liquid-gas systems. In: Perry JH (ed) *Chemical Engineers' Handbook*, McGraw-Hill Book Co, New York
- Tsuge H (1986) Hydrodynamics of bubble formation from submerged orifices. In: Cheremisinoff NP (ed) *Encyclopedia of fluid mechanics*, vol 3. Gulf Publishing Co., Houston
- Várhegyi A, Baranyi I, Somogyi G (1986) A model for the vertical subsurface radon transport in “geogas” microbubbles. *Geophys Trans* 32:235–253
- Walker MI, McKay WA, Pattenden NJ, Liss PS (1986) Actinide enrichment in marine aerosols. *Nature* 323:141–143
- Witherspoon PA, Saraf DN (1965) Diffusion of methane, ethane, propane, and n-Butane in water from 25 to 43. *J Phys Chem* 69:3752–3755