

Transport relations for surface integrals arising in the formulation of balance laws for evolving fluid interfaces

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We establish transport relations for integrals over evolving fluid interfaces. These relations make it possible to localize integral balance laws over non-material interfaces separating fluid phases and, therefore, obtain associated interface conditions in differential form.

1. Introduction

In formulating integral balance laws for a non-material evolving interface $\mathcal{S}(t)$ separating two fluid phases, one often encounters terms of the form

$$\frac{d}{dt} \int_{\mathcal{A}(t)} \varphi(\mathbf{x}, t) da, \quad (1.1)$$

with $\varphi(\mathbf{x}, t)$ a surface field on $\mathcal{S}(t)$, and $\mathcal{A}(t)$ an arbitrary evolving subsurface of $\mathcal{S}(t)$. To obtain the local differential consequences of such laws necessitates an appropriate transport relation. We here establish such relations.

In applications of our results, the surface field under consideration would generally – but not always – represent a *surface excess field* associated with a bulk quantity such as internal energy, entropy, or concentration. There is a large literature on limiting processes leading from bulk fields to surface excess fields (see, e.g. Slattery 1990; Edwards, Brenner & Wasan 1991). Our results are independent of such limiting processes.

To see the difficulties involved in deriving transport relations for (1.1) it is useful to consider the analogous problem associated with the integral

$$\frac{d}{dt} \int_{\mathcal{R}(t)} \Phi(\mathbf{x}, t) dv \quad (1.2)$$

of a bulk scalar-field $\Phi(\mathbf{x}, t)$ over a time-dependent region $\mathcal{R}(t)$ migrating through a fluid. Specifically, assume that the boundary $\partial\mathcal{R}(t)$ moves with (scalar) normal velocity $V_{\partial\mathcal{R}}(\mathbf{x}, t)$ in the direction of its outward unit normal $\mathbf{m}(\mathbf{x}, t)$ and write $V_{\partial\mathcal{R}}^{mig} = V_{\partial\mathcal{R}} - \mathbf{u} \cdot \mathbf{m}$ for the normal velocity of $\partial\mathcal{R}$ relative to the fluid. Two well-known generalizations of the Reynolds (1903) transport relation (cf. Gurtin 1981, p. 78) then

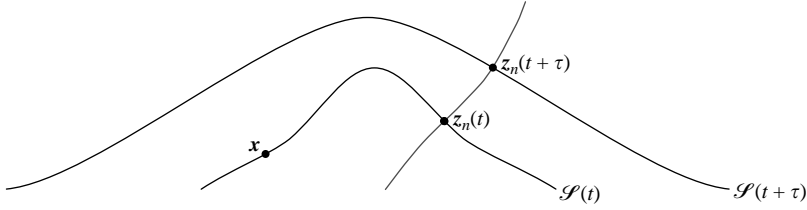


FIGURE 1. Two-dimensional schematic illustrating why a point \mathbf{x} lying on an interface \mathcal{S} at time t need not lie on the interface at a subsequent time $t + \tau$ and, thus, why the partial time derivative of a surface field φ is generally undefined. Also shown is a normal trajectory passing through the points $\mathbf{z}_n(t)$ and $\mathbf{z}_n(t + \tau)$ on $\mathcal{S}(t)$ and $\mathcal{S}(t + \tau)$.

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$$\frac{d}{dt} \int_{\mathcal{R}} \Phi dv = \int_{\mathcal{R}} \left\{ \frac{\partial \Phi}{\partial t} + \text{div}(\Phi \mathbf{u}) \right\} dv + \int_{\partial \mathcal{R}} \Phi V_{\partial R}^{\text{mig}} da, \tag{1.3a}$$

$$\frac{d}{dt} \int_{\mathcal{R}} \Phi dv = \int_{\mathcal{R}} \{ \dot{\Phi} + \Phi \text{div} \mathbf{u} \} dv + \int_{\partial \mathcal{R}} \Phi V_{\partial R}^{\text{mig}} da. \tag{1.3b}$$

Here, $\dot{\Phi}$ (often written $D\Phi/Dt$) denotes the *material time derivative* of Φ , and (1.3b) follows from (1.3a) using the standard identity

$$\dot{\Phi} = \frac{\partial \Phi}{\partial t} + \mathbf{u} \cdot \text{grad} \Phi. \tag{1.4}$$

A difficulty in deriving counterparts for surfaces of the bulk relations (1.3) is associated with determining appropriate superficial analogues of the time derivatives $\dot{\Phi}$ and $\partial \Phi / \partial t$. In this regard, bear in mind that, for φ a surface field, if the surface is not material, then φ is not well-defined: since material points flow across $\mathcal{S}(t)$, it is not generally possible to compute a time derivative holding material points fixed.

Further, while one finds in the literature time derivatives of surface fields φ expressed as conventional partial derivatives $\partial \varphi / \partial t$, such partial derivatives, without explanation, are meaningless: difference quotients of the

$$\frac{\varphi(\mathbf{x}, t + \tau) - \varphi(\mathbf{x}, t)}{\tau} \tag{1.5}$$

are generally undefined because there is no assurance that \mathbf{x} lies on $\mathcal{S}(t + \tau)$ when \mathbf{x} lies on $\mathcal{S}(t)$, even for sufficiently small τ (figure 1). This observation holds even when $\mathcal{S}(t)$ is material. Of course, $\partial \varphi / \partial t$ may be defined using an extension of $\varphi(\mathbf{x}, t)$, at each t , to a three-dimensional region containing the surface; unfortunately, $\partial \varphi / \partial t$ so defined generally depends on the particular extension used.

The main results of this study are surface counterparts of the transport relations (1.3a, b) for the integral (1.1). In particular, the counterpart of (1.3b) requires finding an analogue – for a field φ defined on a surface $\mathcal{S}(t)$ migrating with respect to the material – of the material time derivative $\dot{\Phi}$ of a bulk field Φ . This analogue, which we write as $\overset{\circ}{\varphi}$, turns out to be a time derivative following the evolution of the surface $\mathcal{S}(t)$, a time derivative that accounts for the migration of $\mathcal{S}(t)$ through the fluid. To the best of our knowledge this notion is new; we have already found it useful in developing thermodynamically consistent evolution equations for a fluid–fluid phase interface (Anderson *et al.* 2005).

2. Surfaces

2.1. Gradient and divergence on a surface

Let \mathcal{S} be a surface oriented by a unit-normal field $\mathbf{n}(\mathbf{x})$. A surface field is a field defined on \mathcal{S} . A tangential vector field is a surface vector field $\mathbf{f}(\mathbf{x})$ that satisfies $\mathbf{f} \cdot \mathbf{n} = 0$. We write $\text{grad}_{\mathcal{S}}$ and $\text{div}_{\mathcal{S}}$ for the surface gradient and surface divergence on \mathcal{S} . (We omit smoothness assumptions associated with surfaces, evolving surfaces and surface fields. See McConnell (1957) for definitions of $\text{grad}_{\mathcal{S}}$ and $\text{div}_{\mathcal{S}}$. See, also, Aris (1962).) For φ a scalar surface field.

$$\text{grad}_{\mathcal{S}}\varphi \text{ is a tangential vector field.} \tag{2.1}$$

The field defined by

$$K = -\text{div}_{\mathcal{S}}\mathbf{n} \tag{2.2}$$

is the total (i.e. twice the mean) curvature.

2.2. Surface fields determined by limits of bulk fields

For a bulk field that is well-defined and smooth up to the surface from one or both sides, the surface gradient is simply the tangential component of the standard gradient; e.g. for such a bulk field Φ ,

$$\text{grad}_{\mathcal{S}}\Phi = \text{grad}\Phi - (\mathbf{n} \cdot \text{grad}\Phi)\mathbf{n}. \tag{2.3}$$

(If a bulk field Φ is smooth up to the interface from each side, but not across the interface, then we would have two surface gradients $\text{grad}_{\mathcal{S}}\Phi^{\pm}$, one for each of the limiting values Φ^{\pm} of Φ .)

3. Evolving surfaces

3.1. Local parameterization. Normal velocity

We now consider an evolving surface $\mathcal{S}(t)$ oriented by a unit-normal field $\mathbf{n}(\mathbf{x}, t)$. $\mathcal{S}(t)$ may be parameterized locally – that is, near any time t_0 and point \mathbf{x}_0 on $\mathcal{S}(t_0)$ – by a mapping

$$\mathbf{x} = \hat{\mathbf{x}}(\xi_1, \xi_2, t) \tag{3.1}$$

that, at each time t , establishes a one-to-one correspondence between points (ξ_1, ξ_2) – in an open set in a two-dimensional parameter space – and points \mathbf{x} on $\mathcal{S}(t)$. Writing $(\xi_1, \xi_2) = \hat{\xi}(\mathbf{x}, t)$ for the corresponding inverse map at fixed time, the function

$$\mathbf{v}(\mathbf{x}, t) = \left. \frac{\partial \hat{\mathbf{x}}}{\partial t} \right|_{(\xi_1, \xi_2) = \hat{\xi}(\mathbf{x}, t)} \tag{3.2}$$

represents a local velocity field for $\mathcal{S}(t)$. This velocity field depends on the choice of parameterization: specifically, the normal component of \mathbf{v} , the scalar normal-velocity

$$V = \mathbf{v} \cdot \mathbf{n}, \tag{3.3}$$

is independent of the parameterization, but the tangential velocity is not.

The vectorial counterpart of V is the vector normal-velocity

$$\mathbf{v}_n = V\mathbf{n}. \tag{3.4}$$

3.2. Velocity fields. Trajectories

Given any tangential vector-field $\mathbf{t}(\mathbf{x}, t)$, consider the surface vector field

$$\mathbf{v} \stackrel{\text{def}}{=} V\mathbf{n} + \mathbf{t}.$$

Any such \mathbf{v} represents a velocity field for \mathcal{S} in the sense that there exists a local parameterization (3.1) such that (3.2) holds. Fix $\mathbf{x}_0 \in \mathcal{S}(t_0)$ and write $\mathbf{x}_0 = \hat{\mathbf{x}}(\xi_1^0, \xi_2^0, t_0)$: the curve

$$\mathbf{z}(t) = \hat{\mathbf{x}}(\xi_1^0, \xi_2^0, t) \quad (3.5)$$

is referred to as a trajectory corresponding to the velocity field \mathbf{v} , since

$$\frac{d\mathbf{z}(t)}{dt} = \mathbf{v}(\mathbf{z}(t), t), \quad \mathbf{z}(t_0) = \mathbf{x}_0. \quad (3.6)$$

Trajectories corresponding to the vector normal-velocity \mathbf{v}_n are called normal trajectories (figure 1).

3.3. Time derivatives following $\mathcal{S}(t)$

What we require is a time derivative of a surface scalar field φ following the evolution of the surface $\mathcal{S}(t)$. The simplest such time derivative makes use of the parameterization $\mathbf{x} = \hat{\mathbf{x}}(\xi_1, \xi_2, t)$ of $\mathcal{S}(t)$ and may be defined as follows

$$\frac{\delta\varphi}{\delta t}(\mathbf{x}, t) = \left[\frac{d}{dt} \varphi(\hat{\mathbf{x}}(\xi_1, \xi_2, t), t) \right]_{(\xi_1, \xi_2) = \hat{\xi}(\mathbf{x}, t)}. \quad (3.7)$$

We refer to $\delta\varphi/\delta t$ as the parameter-dependent time derivative following $\mathcal{S}(t)$. The drawback of this time derivative is its dependence on the choice of parameterization.

A time derivative that is independent of the choice of parameterization is the normal time derivative $\overset{\square}{\varphi}$ of φ following \mathcal{S} (Hayes 1957; Thomas 1957). The field $\overset{\square}{\varphi}$ may be defined as follows: choose, arbitrarily, a time t_0 and a point \mathbf{x}_0 on $\mathcal{S}(t_0)$, and let $\mathbf{z}_n(t)$ denote the normal trajectory through \mathbf{x}_0 at t_0 ; then

$$\overset{\square}{\varphi}(\mathbf{x}_0, t_0) = \left[\frac{d}{dt} \varphi(\mathbf{z}_n(t), t) \right]_{t=t_0}. \quad (3.8)$$

3.4. Normally constant extension of a surface field

We can also compute the time derivative of a surface field using an extension of the field to a three-dimensional neighbourhood of the surface, but such a time derivative depends on the extension used. A geometrically natural method of smoothly extending a surface field $\varphi(\mathbf{x}, t)$, at each time, to a three-dimensional region containing the surface is obtained by requiring that φ be constant on normal lines, where a normal line at time t is a line through a point \mathbf{x} on $\mathcal{S}(t)$ parallel to $\mathbf{n}(\mathbf{x}, t)$. The extension $\hat{\varphi}$ obtained in this manner is referred to as a normally constant extension of φ . (Since normal lines may cross, such an extension is generally valid, at each t , at most in a neighbourhood of $\mathcal{S}(t)$.)

We now relate the partial time derivative of the field $\hat{\varphi}$ to the normal time derivative of φ . Bearing in mind the right-hand side of (3.8), the chain rule yields

$$\frac{d}{dt} \varphi(\mathbf{z}_n(t), t) = \frac{d}{dt} \hat{\varphi}(\mathbf{z}_n(t), t) = \left[\frac{\partial \hat{\varphi}(\mathbf{x}, t)}{\partial t} + \mathbf{v}_n(\mathbf{x}, t) \cdot \text{grad} \hat{\varphi}(\mathbf{x}, t) \right]_{\mathbf{x} = \mathbf{z}_n(t)}. \quad (3.9)$$

Next, since $\hat{\varphi}$ is constant on normal lines, for points on the surface,

$$\mathbf{n} \cdot \text{grad} \hat{\varphi} = 0,$$

so that, by (2.3),

$$\text{grad}_{\mathcal{S}} \varphi = \text{grad} \hat{\varphi}. \quad (3.10)$$

Further, $\mathbf{v}_n \cdot \text{grad}_{\mathcal{S}}\varphi = V\mathbf{n} \cdot \text{grad}_{\mathcal{S}}\varphi = 0$, so that, by (3.8), (3.9) and (3.10),

$$\bar{\varphi} = \frac{\partial \hat{\varphi}}{\partial t}. \tag{3.11}$$

Thus the normal time derivative is the conventional partial time derivative of φ when φ is extended to be constant on normal lines.

Our next step is to determine the relation of $\partial \hat{\varphi} / \partial t$ to the time derivative $\delta \varphi / \delta t$ defined using the parameterization $\mathbf{x} = \hat{\mathbf{x}}(\xi_1, \xi_2, t)$. Let \mathbf{v} be the velocity (3.2) associated with this parameterization. Then, focusing on the right-hand side of (3.7) we find, using the chain rule, that

$$\frac{d}{dt} \varphi(\hat{\mathbf{x}}(\xi_1, \xi_2, t), t) = \left[\frac{\partial \hat{\varphi}(\mathbf{x}, t)}{\partial t} + \mathbf{v}(\mathbf{x}, t) \cdot \text{grad} \hat{\varphi}(\mathbf{x}, t) \right]_{\mathbf{x} = \hat{\mathbf{x}}(\xi_1, \xi_2, t)}.$$

But, by (3.10), for $\mathbf{x} = \hat{\mathbf{x}}(\xi_1, \xi_2, t)$,

$$\mathbf{v}(\mathbf{x}, t) \cdot \text{grad} \hat{\varphi}(\mathbf{x}, t) = \mathbf{v}(\mathbf{x}, t) \cdot \text{grad}_{\mathcal{S}}\varphi(\mathbf{x}, t) = \mathbf{v}_{tan}(\mathbf{x}, t) \cdot \text{grad}_{\mathcal{S}}\varphi(\mathbf{x}, t).$$

Thus, by (3.7) and (3.11), we obtain a simple relation,

$$\bar{\varphi} = \frac{\delta \varphi}{\delta t} - \mathbf{v}_{tan} \cdot \text{grad}_{\mathcal{S}}\varphi, \tag{3.12}$$

that allows us to calculate the normal time derivative, given the parameter-dependent time derivative.

3.5. Basic transport relation for a surface integrals

Consider an arbitrary evolving subsurface $\mathcal{A}(t)$ of $\mathcal{S}(t)$ with boundary curve $\partial \mathcal{A}(t)$ oriented by its exterior unit-normal field $\mathbf{v}(\mathbf{x}, t)$; \mathbf{v} is normal to $\partial \mathcal{A}$, but tangent to \mathcal{S} . The curve $\partial \mathcal{A}(t)$ evolves through space, and its motion is described by a velocity field $\mathbf{v}_{\partial \mathcal{A}}(\mathbf{x}, t)$ with \mathbf{x} on $\partial \mathcal{A}(t)$. Only the component $\mathbf{v}_{\partial \mathcal{A}} \cdot \mathbf{v}$ of $\mathbf{v}_{\partial \mathcal{A}}$ normal to $\partial \mathcal{A}$ is independent of the parameterization of $\partial \mathcal{A}$ and, hence, intrinsic to the motion. On the other hand, since $\partial \mathcal{A}(t)$ is a subset of $\mathcal{S}(t)$ for all t , $\mathbf{v}_{\partial \mathcal{A}} \cdot \mathbf{n} = V$. Writing $V_{\partial \mathcal{A}} = \mathbf{v}_{\partial \mathcal{A}} \cdot \mathbf{v}$, we may therefore express the intrinsic component of every velocity field for $\partial \mathcal{A}$ in the form

$$V\mathbf{n} + V_{\partial \mathcal{A}}\mathbf{v}. \tag{3.13}$$

We refer to $V_{\partial \mathcal{A}}(\mathbf{x}, t)$ as the scalar normal-velocity of $\partial \mathcal{A}(t)$; the field $V_{\partial \mathcal{A}}(\mathbf{x}, t)$ describes the intrinsic instantaneous motion of $\partial \mathcal{A}(t)$ on the tangent plane to $\mathcal{S}(t)$ at \mathbf{x} .

As a starting point in developing counterparts, for surfaces, of the Reynolds relations (1.3), we state the following result: given any scalar surface field $\varphi(\mathbf{x}, t)$ and any evolving subsurface $\mathcal{A}(t)$ of $\mathcal{S}(t)$,

$$\frac{d}{dt} \int_{\mathcal{A}} \varphi \, da = \int_{\mathcal{A}} \{ \bar{\varphi} - \varphi K V \} \, da + \int_{\partial \mathcal{A}} \varphi V_{\partial \mathcal{A}} \, ds, \tag{3.14}$$

with $\bar{\varphi}$ the normal time derivative of φ following \mathcal{S} . (This result was established independently by Petryk & Mroz (1986) and Gurtin, Struthers & Williams (1989); see also Estrada & Kanwal (1991). A simple derivation of (3.14) for curves evolving in a planar domain is given by Angenent & Gurtin (1991).)

Using the identity (3.12), this transport relation can be expressed in terms of the parameter-dependent time derivative.

4. Migrating surfaces in fluids

While valid for a surface migrating through a fluid, the transport relation (3.14) is peculiar in that it exhibits no influence of the flow field. We turn now to deriving alternative versions of (3.14) that account for that influence. In this regard, bear in mind that, as noted in §1, for φ a surface field, if the surface is not material, then neither the material time derivative $\dot{\varphi}$ nor the partial derivative $\partial\varphi/\partial t$ is well-defined.

4.1. *Fluid velocity. Migrational velocities*

We now suppose that the evolving surface $\mathcal{S}(t)$ is migrating through a fluid. We write $\mathbf{u}(\mathbf{x}, t)$ for the velocity of the fluid and assume that this velocity has limiting values $\mathbf{u}^+(\mathbf{x}, t)$ and $\mathbf{u}^-(\mathbf{x}, t)$ on each side of $\mathcal{S}(t)$, where \mathbf{u}^+ denotes the limiting value from that side of \mathcal{S} into which \mathbf{n} points. We assume also that the tangential component \mathbf{u}_{tan} of \mathbf{u} is continuous across \mathcal{S} , so that

$$\mathbf{u}^+ - \mathbf{u}^- = (\mathbf{u}^+ \cdot \mathbf{n} - \mathbf{u}^- \cdot \mathbf{n})\mathbf{n}. \tag{4.1}$$

We continue to write $V(\mathbf{x}, t)$ and $\mathbf{v}_n(\mathbf{x}, t)$ for the scalar and vector normal-velocities for $\mathcal{S}(t)$. In addition, we let $\mathbf{v}(\mathbf{x}, t)$ denote a (for now arbitrary) velocity field for $\mathcal{S}(t)$. Then the fields

$$\mathbf{v} - \mathbf{u}^\pm \tag{4.2}$$

represent migrational velocities of \mathcal{S} relative to the fluid material on each of its sides.

Consider an arbitrary migrating subsurface $\mathcal{A}(t)$ of $\mathcal{S}(t)$. The scalar normal velocity $V_{\partial\mathcal{A}}$ of $\partial\mathcal{A}$ in the direction of its unit normal \mathbf{v} is as discussed in the paragraph containing (3.13). Bearing in mind that \mathbf{v} is tangential to \mathcal{S} ,

$$\mathbf{u}^+ \cdot \mathbf{v} = \mathbf{u}^- \cdot \mathbf{v} = \mathbf{u}_{tan} \cdot \mathbf{v}, \tag{4.3}$$

and hence

$$V_{\partial\mathcal{A}}^{mig} \stackrel{\text{def}}{=} V_{\partial\mathcal{A}} - \mathbf{u}^\pm \cdot \mathbf{v} = V_{\partial\mathcal{A}} - \mathbf{u}_{tan} \cdot \mathbf{v} \tag{4.4}$$

represents the normal migrational velocity of $\partial\mathcal{A}$; that is, the normal velocity of $\partial\mathcal{A}$ relative to the fluid.

4.2. *The MB transport relation for a surface migrating through a fluid*

A transport relation for a scalar surface field $\varphi(\mathbf{x}, t)$ that accounts for the fluid may be derived from (3.14). Using the surface divergence theorem,

$$\int_{\partial\mathcal{A}} (\varphi \mathbf{u}_{tan}) \cdot \mathbf{v} \, ds = \int_{\mathcal{A}} \text{div}_{\mathcal{S}}(\varphi \mathbf{u}_{tan}) \, da, \tag{4.5}$$

and this relation, (3.14), and (4.4) together yield the *MB transport relation*:

$$\frac{d}{dt} \int_{\mathcal{A}} \varphi \, da = \int_{\mathcal{A}} \{ \overset{\square}{\varphi} + \text{div}_{\mathcal{S}}(\varphi \mathbf{u}_{tan}) - \varphi K V \} \, da + \int_{\partial\mathcal{A}} \varphi V_{\partial\mathcal{A}}^{mig} \, ds. \tag{4.6}$$

(This result is based on ideas of Mavrouniotis & Brenner (1993), whose derivation of an integral balance for a surface excess field contains the essence of (4.6) (cf. Mavrouniotis 1989; Edwards *et al.* 1991).)

The time derivative $\overset{\square}{\varphi}$ used in the MB transport relation is the time derivative following the surface $\mathcal{S}(t)$ as it evolves through Euclidean space; as such, $\overset{\square}{\varphi}$ does not account for the actual migration of $\mathcal{S}(t)$ through the fluid. In this regard, $\overset{\square}{\varphi}$ is analogous to the spatial time derivative $\partial\Phi(\mathbf{x}, t)/\partial t$ of a field defined in the bulk fluid (cf. (1.4)).

In the next section we introduce a time derivative that accounts explicitly for the migration of $\mathcal{S}(t)$ through the fluid.

4.3. *Migratorially normal time derivative following $\mathcal{S}(t)$*

Our first step is to find a velocity field \mathbf{v} for \mathcal{S} that accounts for the migration of \mathcal{S} through the fluid. Bearing in mind that there are two migrational velocities $\mathbf{v} - \mathbf{u}^\pm$, we now show that we can find a velocity field \mathbf{v} for \mathcal{S} that renders each of the velocities $\mathbf{v} - \mathbf{u}^\pm$ normal. In this regard, note that

$$\mathbf{v} - \mathbf{u}^\pm = \mathbf{v} - (\mathbf{u}^\pm \cdot \mathbf{n})\mathbf{n} - \mathbf{u}_{tan} = (V - \mathbf{u}^\pm \cdot \mathbf{n})\mathbf{n} + (\mathbf{v}_{tan} - \mathbf{u}_{tan}),$$

so that, taking \mathbf{v}_{tan} , which is arbitrary, equal to \mathbf{u}_{tan} , we arrive at a choice $\mathbf{v} = \mathbf{v}^*$ of velocity field for \mathcal{S} with each of its migrational velocities $\mathbf{v}^* - \mathbf{u}^\pm$ normal:

$$\mathbf{v}^* - \mathbf{u}^\pm = (V - \mathbf{u}^\pm \cdot \mathbf{n})\mathbf{n}. \tag{4.7}$$

The resulting velocity field \mathbf{v}^* , called the migratorially normal velocity field for \mathcal{S} , has the specific form

$$\mathbf{v}^* = V\mathbf{n} + \mathbf{u}_{tan} \tag{4.8}$$

and is important because it is normal when computed relative to the material on either side of $\mathcal{S}(t)$.

The migratorially normal time derivative of $\varphi(\mathbf{x}, t)$ following $\mathcal{S}(t)$ is defined – at an arbitrary time t_0 and point \mathbf{x}_0 on $\mathcal{S}(t_0)$ – as follows:

$$\overset{\circ}{\varphi}(\mathbf{x}_0, t_0) = \left[\frac{d}{dt} \varphi(\mathbf{z}^*(t), t) \right]_{t=t_0}, \tag{4.9}$$

where $\mathbf{z}^*(t)$ is the trajectory through \mathbf{x}_0 at time t_0 corresponding to the migratorially normal velocity-field $\mathbf{v}^* = V\mathbf{n} + \mathbf{u}_{tan}$ (cf. the paragraph containing (3.6)).

4.4. *Transport relation based on the migratorially normal time derivative*

In this section, we derive a transport relation for a scalar surface-field $\varphi(\mathbf{x}, t)$ that accounts explicitly for the migration of \mathcal{S} through the field.

Let $\hat{\varphi}$ denote the normally constant extension of φ as defined in §3.4. Since the velocity field underlying the definition of $\overset{\circ}{\varphi}$ is the migratorially normal field $\mathbf{v}^* = V\mathbf{n} + \mathbf{u}_{tan}$, we find, using (3.10), (4.9), and the chain rule, that

$$\overset{\circ}{\varphi} = \mathbf{v}_{tan}^* \cdot \text{grad}_{\mathcal{S}} \varphi + \frac{\partial \hat{\varphi}}{\partial t}. \tag{4.10}$$

Thus, by (3.11), the time derivatives $\overset{\circ}{\varphi}$ and $\overset{\square}{\varphi}$ are related through the important identity

$$\overset{\circ}{\varphi} = \overset{\square}{\varphi} + \mathbf{u}_{tan} \cdot \text{grad}_{\mathcal{S}} \varphi. \tag{4.11}$$

By (4.8) and (4.11),

$$\overset{\square}{\varphi} + \text{div}_{\mathcal{S}}(\varphi \mathbf{u}_{tan}) = \overset{\circ}{\varphi} - \mathbf{u}_{tan} \cdot \text{grad}_{\mathcal{S}} \varphi + \text{div}_{\mathcal{S}}(\varphi \mathbf{u}_{tan}) = \overset{\circ}{\varphi} + \varphi \text{div}_{\mathcal{S}} \mathbf{u}_{tan},$$

and this result together with (4.6) yields our main result, the transport relation

$$\frac{d}{dt} \int_{\mathcal{A}} \varphi \, da = \int_{\mathcal{A}} \{ \overset{\circ}{\varphi} + \varphi \text{div}_{\mathcal{S}} \mathbf{u}_{tan} - \varphi KV \} \, da + \int_{\partial \mathcal{A}} \varphi V_{\partial \mathcal{A}}^{mig} \, ds. \tag{4.12}$$

5. **Material surfaces**

5.1. *Kinemematical relations*

Assume that $\mathcal{S}(t)$ is a material surface so that, necessarily, the fluid velocity is continuous across $\mathcal{S}(t)$. Assume further that $\mathcal{A}(t)$ is a material subsurface of $\mathcal{S}(t)$,

so that the boundary curve $\partial\mathcal{A}(t)$ is a material curve (stated differently: \mathcal{S} , \mathcal{A} , and $\partial\mathcal{A}$ convect with the fluid). Then:

(i) The fluid velocity \mathbf{u} is a velocity field for \mathcal{S} ; hence the normal velocity of \mathcal{S} and the normal fluid-velocity coincide,

$$V = \mathbf{u} \cdot \mathbf{n}. \tag{5.1}$$

(ii) The migrationally normal velocity field for \mathcal{S} coincides with the fluid velocity,

$$\mathbf{u} = V\mathbf{n} + \mathbf{u}_{tan}. \tag{5.2}$$

(iii) The material time derivative $\dot{\varphi}$ coincides with the time derivative $\overset{\circ}{\varphi}$ following the surface as described by the migrationally normal velocity field (4.8),

$$\dot{\varphi} = \overset{\circ}{\varphi}. \tag{5.3}$$

(iv) The normal migrational velocity $V_{\partial\mathcal{A}}^{mig}$ vanishes. Assertion (i) is immediate, as is the relation

$$V_{\partial\mathcal{A}} = \mathbf{u} \cdot \mathbf{v}, \tag{5.4}$$

which implies (iv). By (i),

$$\mathbf{v} = V\mathbf{n} + \mathbf{u}_{tan} = (\mathbf{u} \cdot \mathbf{n})\mathbf{n} + \mathbf{u}_{tan} = \mathbf{u},$$

which is (ii). Finally, by (ii) and (3.6), the trajectories used to compute (4.9) satisfy

$$\frac{dz(t)}{dt} = \mathbf{u}(z(t), t),$$

and hence represent trajectories of material points. Thus, (iii) is satisfied.

5.2. Transport relations for material surfaces

The following transport relations follow as consequences of (4.12): if $\mathcal{S}(t)$ is a material surface and $\mathcal{A}(t)$ a material subsurface of $\mathcal{S}(t)$, with boundary curve $\partial\mathcal{A}(t)$ a material curve, then given any scalar surface field $\varphi(\mathbf{x}, t)$,

$$\frac{d}{dt} \int_{\mathcal{A}} \varphi \, da = \int_{\mathcal{A}} \{ \dot{\varphi} + \varphi \operatorname{div}_{\mathcal{S}} \mathbf{u}_{tan} - \varphi (\mathbf{u} \cdot \mathbf{n}) K \} \, da, \tag{5.5a}$$

$$\frac{d}{dt} \int_{\mathcal{A}} \varphi \, da = \int_{\mathcal{A}} \{ \dot{\varphi} + \varphi \operatorname{div}_{\mathcal{S}} \mathbf{u} \} \, da. \tag{5.5b}$$

Equation (5.5a) follows directly upon using (5.1) and (5.3) in (4.12); this equation was previously established by Slattery (1972) (cf. Slattery 1990, equation (3–6)). To establish (5.5b), note that, by (2.2),

$$-(\mathbf{u} \cdot \mathbf{n})K = (\mathbf{u} \cdot \mathbf{n})\operatorname{div}_{\mathcal{S}} \mathbf{n} = \operatorname{div}_{\mathcal{S}} ((\mathbf{u} \cdot \mathbf{n})\mathbf{n}) - \underbrace{\mathbf{n} \cdot \operatorname{grad}_{\mathcal{S}} (\mathbf{u} \cdot \mathbf{n})}_{=0},$$

so that

$$-(\mathbf{u} \cdot \mathbf{n})K + \operatorname{div}_{\mathcal{S}} \mathbf{u}_{tan} = \operatorname{div}_{\mathcal{S}} \mathbf{u},$$

and (5.5a) reduces to (5.5b).

Transport relations similar to (5.5), but somewhat more general, hold when $\mathcal{S}(t)$ is material, but $\mathcal{A}(t)$ is non-material, so that $V_{\partial\mathcal{A}}^{mig} \neq 0$. These relations have the form

$$\left. \begin{aligned} \frac{d}{dt} \int_{\mathcal{A}} \varphi \, da &= \int_{\mathcal{A}} \{ \dot{\varphi} + \varphi \operatorname{div}_{\mathcal{S}} \mathbf{u}_{tan} - \varphi (\mathbf{u} \cdot \mathbf{n}) K \} \, da + \int_{\partial\mathcal{A}} \varphi V_{\partial\mathcal{A}}^{mig} \, ds, \\ \frac{d}{dt} \int_{\mathcal{A}} \varphi \, da &= \int_{\mathcal{A}} \{ \dot{\varphi} + \varphi \operatorname{div}_{\mathcal{S}} \mathbf{u} \} \, da + \int_{\partial\mathcal{A}} \varphi V_{\partial\mathcal{A}}^{mig} \, ds. \end{aligned} \right\} \quad (5.6)$$

6. Comparisons

It is interesting to compare the transport relations of Reynolds for a region $\mathcal{R}(t)$ evolving through a fluid to those relations derived here for a surface $\mathcal{S}(t)$ evolving through a fluid. In particular, the Reynolds relation (1.3a) should be compared to the MB relation (4.6),

$$\begin{aligned} \frac{d}{dt} \int_{\mathcal{R}} \Phi \, dv &= \int_{\mathcal{R}} \left\{ \frac{\partial \Phi}{\partial t} + \operatorname{div}(\Phi \mathbf{u}) \right\} \, dv + \int_{\partial\mathcal{R}} \Phi V_{\partial\mathcal{R}}^{mig} \, da, \\ \frac{d}{dt} \int_{\mathcal{A}} \varphi \, da &= \int_{\mathcal{A}} \{ \overset{\square}{\varphi} + \operatorname{div}_{\mathcal{S}}(\varphi \mathbf{u}_{tan}) - \varphi K V \} \, da + \int_{\partial\mathcal{A}} \varphi V_{\partial\mathcal{A}}^{mig} \, ds, \end{aligned}$$

while the Reynolds relation (1.3b) should be compared to the relation (4.12),

$$\begin{aligned} \frac{d}{dt} \int_{\mathcal{R}} \Phi \, dv &= \int_{\mathcal{R}} \{ \dot{\Phi} + \Phi \operatorname{div} \mathbf{u} \} \, dv + \int_{\partial\mathcal{R}} \Phi V_{\partial\mathcal{R}}^{mig} \, da, \\ \frac{d}{dt} \int_{\mathcal{A}} \varphi \, da &= \int_{\mathcal{A}} \{ \overset{\circ}{\varphi} + \varphi \operatorname{div}_{\mathcal{S}} \mathbf{u}_{tan} - \varphi K V \} \, da + \int_{\partial\mathcal{A}} \varphi V_{\partial\mathcal{A}}^{mig} \, ds. \end{aligned}$$

On using an arrow \rightarrow to indicate the relation ‘is analogous to’, these comparisons would seem to suggest that

$$\frac{\partial \Phi}{\partial t} \rightarrow \overset{\square}{\varphi}, \quad \dot{\Phi} \rightarrow \overset{\circ}{\varphi}, \quad \mathbf{u} \rightarrow \mathbf{u}_{tan}. \quad (6.1)$$

These analogies are consistent with the identity $\dot{\varphi} = \overset{\circ}{\varphi}$ for a material surface (cf. (5.3)) and with a comparison between the relations (1.4) and (4.11):

$$\dot{\varphi} = \frac{\partial \Phi}{\partial t} + \mathbf{u} \cdot \operatorname{grad} \Phi, \quad \overset{\circ}{\varphi} = \overset{\square}{\varphi} + \mathbf{u}_{tan} \cdot \operatorname{grad}_{\mathcal{S}} \varphi.$$

When discussing the bulk motion of a fluid, the material time derivative $\dot{\Phi}$ of a bulk field Φ embodies more of the physics associated with the actual motion of material particles than does $\partial\Phi/\partial t$. Analogously, when discussing an interface migrating through a fluid, the migrationally normal time derivative $\overset{\circ}{\varphi}$ of an interfacial field φ embodies more of the physics associated with the motion of the interface relative to the fluid than does the normal time derivative $\overset{\square}{\varphi}$. We therefore expect the migrationally normal time derivative to be particularly valuable in the study of theoretical issues, such as the derivation of interface conditions from general thermomechanical principles (cf. Anderson *et al.* (2005), who develop interface conditions for phase transformations in viscous heat-conducting fluids; there, the underlying physics leads naturally to interfacial expressions of the first two laws of thermodynamics in terms of migrationally normal time derivatives). On the other hand, for numerical investigations, the normal time derivative might be more directly implemented (much as the partial time derivative is more directly implemented than the material time derivative in simulations of bulk fluid flow).

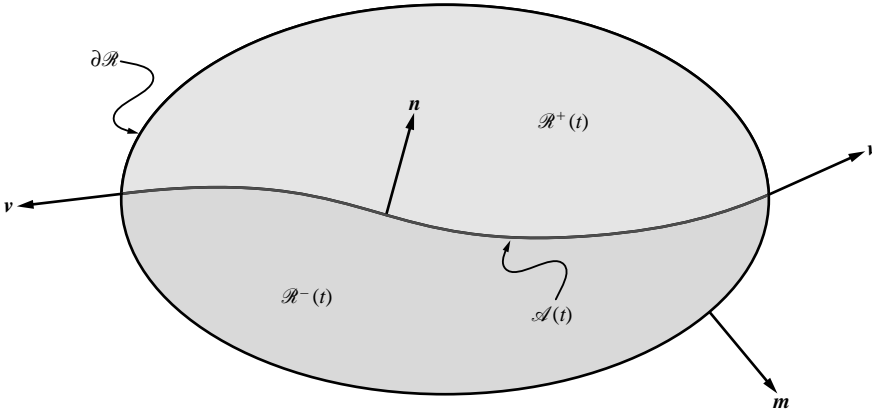


FIGURE 2. Two-dimensional schematic illustrating a control volume \mathcal{R} divided by a portion $\mathcal{A}(t)$ of the interface $\mathcal{S}(t)$ into time-dependent subsets $\mathcal{R}^+(t)$ and $\mathcal{R}^-(t)$. Whereas \mathbf{m} is the unit normal on $\partial\mathcal{R}$, directed outward from \mathcal{R} , \mathbf{n} is the unit normal on \mathcal{S} , directed into \mathcal{R}^+ and \mathbf{v} is the unit normal on $\partial\mathcal{A}$, directed outward from \mathcal{A} .

7. Application to solute transport

Consider the flow of a two-phase binary solution with phases separated by the interface $\mathcal{S}(t)$. Label the phases \pm with the (+) phase the phase into which \mathbf{n} points. Let c and \mathbf{j} denote the concentration and flux of the solute in bulk, Γ and \mathbf{h} the surface excess concentration and flux of the solute on \mathcal{S} . Assume that the bulk fields c and \mathbf{j} are smooth up to the interface from either side, but suffer jump discontinuities across the interface and let $[[c]] = c^+ - c^-$ and $[[\mathbf{j}]] = \mathbf{j}^+ - \mathbf{j}^-$.

Let \mathcal{R} denote a fixed control volume through which the fluid flows and assume that \mathcal{R} intersects the interface in a smooth subsurface $\mathcal{A}(t)$. Let \mathbf{m} denote the outward unit normal to $\partial\mathcal{R}$ and \mathbf{v} the outward unit normal to $\partial\mathcal{A}$. Then, balance of solute mass in \mathcal{R} requires that, at each time t ,

$$\frac{d}{dt} \left\{ \int_{\mathcal{R}} c \, dv + \int_{\mathcal{A}} \Gamma \, da \right\} = - \int_{\partial\mathcal{R}} \{ \mathbf{j} \cdot \mathbf{m} + c \mathbf{u} \cdot \mathbf{m} \} \, da - \int_{\partial\mathcal{A}} \{ \mathbf{h} \cdot \mathbf{v} - \Gamma V_{\mathcal{A}}^{mig} \} \, ds. \quad (7.1)$$

We now localize (7.1) to the interface. Note first that, letting \mathcal{R}^\pm and $(\partial\mathcal{R})^\pm$ denote the portions of \mathcal{R} and $\partial\mathcal{R}$ in the (\pm) phase (so that neither of $(\partial\mathcal{R})^\pm$ contains interior points of \mathcal{A}), we may use a standard transport theorem to verify that, since \mathcal{R} is a fixed region,

$$\frac{d}{dt} \int_{\mathcal{R}^+} c \, dv = \int_{\mathcal{R}^+} \frac{\partial c}{\partial t} \, dv - \int_{\mathcal{A}} c V \, da, \quad \frac{d}{dt} \int_{\mathcal{R}^-} c \, dv = \int_{\mathcal{R}^-} \frac{\partial c}{\partial t} \, dv + \int_{\mathcal{A}} c V \, da.$$

Thus, bearing in mind figure 2, we find that, in the limit as \mathcal{R} shrinks to the interface,

$$\int_{\mathcal{R}^\pm} \frac{\partial c}{\partial t} \, dv \rightarrow 0, \quad \int_{(\partial\mathcal{R})^\pm} \mathbf{j} \cdot \mathbf{m} \, da \rightarrow \pm \int_{\mathcal{A}} \mathbf{j}^\pm \cdot \mathbf{n} \, da \quad \int_{(\partial\mathcal{R})^\pm} c \mathbf{u} \cdot \mathbf{m} \, da \rightarrow \pm \int_{\mathcal{A}} c^\pm \mathbf{u}^\pm \cdot \mathbf{n} \, da,$$

and (7.1) yields

$$\frac{d}{dt} \int_{\mathcal{A}} \Gamma \, da = - \int_{\partial\mathcal{A}} \{ \mathbf{h} \cdot \mathbf{v} - \Gamma V_{\mathcal{A}}^{mig} \} \, ds - \int_{\mathcal{A}} \{ [[\mathbf{j}]] \cdot \mathbf{n} - [[c]](V - \mathbf{u} \cdot \mathbf{n}) \} \, da. \quad (7.2)$$

Thus, applying the surface divergence theorem to the term involving \mathbf{h} , using the transport relation (4.12), and dividing by the area of \mathcal{A} , we arrive at

$$\frac{1}{\text{area}(\mathcal{A})} \int_{\mathcal{A}} \{\overset{\circ}{\Gamma} + \Gamma \text{div}_{\mathcal{S}} \mathbf{u}_{tan} - \Gamma K V + \text{div}_{\mathcal{S}} \mathbf{h} + \llbracket \mathbf{j} \rrbracket \cdot \mathbf{n} - \llbracket c(V - \mathbf{u} \cdot \mathbf{n}) \rrbracket\} da = 0. \quad (7.3)$$

Since the region \mathcal{R} containing \mathcal{A} was arbitrarily chosen, we may assume that, given an arbitrary time t_0 and an arbitrary point \mathbf{x}_0 that lies on $\mathcal{S}(t_0)$, the subsurface $\mathcal{A}(t_0)$ contains \mathbf{x}_0 and has area arbitrarily small. Thus, passing to the limit as $\text{area}(\mathcal{A}(t_0)) \rightarrow 0$ – and using the fact that t_0 and \mathbf{x}_0 on $\mathcal{S}(t_0)$ were arbitrarily chosen – we arrive at the relation

$$\overset{\circ}{\Gamma} + \Gamma \text{div}_{\mathcal{S}} \mathbf{u}_{tan} - \Gamma K V + \text{div}_{\mathcal{S}} \mathbf{h} = -\llbracket \mathbf{j} \rrbracket \cdot \mathbf{n} + \llbracket c(V - \mathbf{u} \cdot \mathbf{n}) \rrbracket, \quad (7.4)$$

valid pointwise on the interface. Equivalently, using (4.11), we arrive at a balance

$$\overset{\square}{\Gamma} + \text{div}_{\mathcal{S}}(\Gamma \mathbf{u}_{tan}) - \Gamma K V + \text{div}_{\mathcal{S}} \mathbf{h} = -\llbracket \mathbf{j} \rrbracket \cdot \mathbf{n} + \llbracket c(V - \mathbf{u} \cdot \mathbf{n}) \rrbracket \quad (7.5)$$

due to Mavrouniotis & Brenner (1993).

When \mathcal{S} is a material surface, then $\overset{\circ}{\Gamma} = \dot{\Gamma}$, $V = \mathbf{u} \cdot \mathbf{n}$, and the balance (7.4) becomes

$$\dot{\Gamma} + \Gamma \text{div}_{\mathcal{S}} \mathbf{u}_{tan} - \Gamma K \mathbf{u} \cdot \mathbf{n} + \text{div}_{\mathcal{S}} \mathbf{h} = -\llbracket \mathbf{j} \rrbracket \cdot \mathbf{n}, \quad (7.6)$$

which is consistent with equations presented by Scriven (1960), Aris (1962), Slattery (1972), Waxman (1984), and Stone (1989) provided, as noted by Edwards *et al.* (1991), one considers $\partial \Gamma / \partial t$ as given by

$$\frac{\partial \Gamma}{\partial t} = \dot{\Gamma} - \mathbf{u}_{tan} \cdot \text{grad}_{\mathcal{S}} \Gamma, \quad (7.7)$$

a relation that holds automatically when $\partial \Gamma / \partial t$ is defined as the normal time derivative. (Cf. Wong, Rumschitzki & Maldarelli (1996), who use the normally constant extension to define the partial time derivative of this extension on the interface, as well as the numerical schemes of Stone & Leal (1990), Borhan & Mao (1992), and Milliken, Stone & Leal (1993), which make tacit use of the normally constant extension.)

Next, we apply the general balance (7.4) to the topic of evaporating surfactant solutions assuming, as do Danov *et al.* (1998), that surfactant evaporation is negligible. Then on letting the (+) phase denote the vapour, both c^+ and \mathbf{j}^+ vanish and (7.4) becomes

$$\overset{\circ}{\Gamma} + \Gamma \text{div}_{\mathcal{S}} \mathbf{u}_{tan} - \Gamma K V + \text{div}_{\mathcal{S}} \mathbf{h} = \mathbf{j}^- \cdot \mathbf{n} - c^-(V - \mathbf{u}^- \cdot \mathbf{n}). \quad (7.8)$$

Here, we note that previous statements of the surfactant balance on the solution surface have been in error. In particular, consider equation (3b) of Danov *et al.* (1998) To clarify the comparison between (7.8) and that equation, suppose that the bulk and surface fluxes are given by $\mathbf{j} = -D \text{grad} c$ and $\mathbf{h} = -D_{\mathcal{S}} \text{grad}_{\mathcal{S}} \Gamma$, in which case, (7.8) specializes to

$$\overset{\circ}{\Gamma} + \Gamma \text{div}_{\mathcal{S}} \mathbf{u}_{tan} - \Gamma K V = \text{div}_{\mathcal{S}}(D_{\mathcal{S}} \text{grad}_{\mathcal{S}} \Gamma) - \mathbf{n} \cdot (D \text{grad} c)^- - c^-(V - \mathbf{u}^- \cdot \mathbf{n}). \quad (7.9)$$

In place of the left-hand side of (7.9), Danov *et al.* (1998) write

$$\frac{\partial \Gamma}{\partial t} + \text{div}_{\mathcal{S}}(\Gamma \mathbf{u}_{tan}), \quad (7.10)$$

without specifying the meaning of the partial time derivative $\partial\Gamma/\partial t$. If we use the normally constant extension of Γ , then

$$\frac{\partial\Gamma}{\partial t} = \overset{\circ}{\Gamma} - \mathbf{u}_{tan} \cdot \text{grad}_{\mathcal{S}}\Gamma, \quad (7.11)$$

and (7.10) coincides with the first two terms on the left-hand side of (7.9). Even then, however, equation (3b) of Danov *et al.* (1998) is missing the term $-\Gamma KV$ on its left-hand side. Moreover, whereas the definition (7.7) arising for a material surface can be deduced without recourse to the notion of the normal time derivative, (7.11) requires consideration of that derivative (or the migrationally normal time derivative) and is therefore non-trivial.

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