REVIEW ARTICLE



# **A review of solid–fuid selection options for optical‑based measurements in single‑phase liquid, two‑phase liquid–liquid and multiphase solid–liquid fows**

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**Abstract** Experimental techniques based on optical measurement principles have experienced signifcant growth in recent decades. They are able to provide detailed information with high-spatiotemporal resolution on important scalar (e.g., temperature, concentration, and phase) and vector (e.g., velocity) felds in single-phase or multiphase fows, as well as interfacial characteristics in the latter, which has been instrumental to step-changes in our fundamental understanding of these fows, and the development and validation of advanced models with ever-improving predictive accuracy and reliability. Relevant techniques rely upon well-established optical methods such as direct photography, laser-induced fuorescence, laser Doppler velocimetry/ phase Doppler anemometry, particle image/tracking velocimetry, and variants thereof. The accuracy of the resulting data depends on numerous factors including, importantly, the refractive indices of the solids and liquids used. The best results are obtained when the observational materials have closely matched refractive indices, including testsection walls, liquid phases, and any suspended particles. This paper reviews solid–liquid and solid–liquid–liquid refractive-index-matched systems employed in different felds, e.g., multiphase fows, turbomachinery, bio-fuid flows, with an emphasis on liquid–liquid systems. The refractive indices of various aqueous and organic phases found in the literature span the range 1.330–1.620 and 1.251–1.637, respectively, allowing the identifcation of appropriate combinations to match selected transparent or translucent plastics/polymers, glasses, or custom materials in single-phase liquid or multiphase liquid–liquid fow systems. In addition, the refractive indices of fuids can be further tuned with the use of additives, which also allows for the matching of important fow similarity parameters such as density and viscosity.

### **1 Introduction**

This paper reviews the practice of refractive-index matching (RIM) in experimental work concerned with at least one liquid as the base fuid and excluding systems with a gaseous phase, thus focusing on solid–liquid and liquid– liquid combinations for use in two-phase liquid–liquid and multiphase solid–liquid fows, while retaining applicability to single-phase liquid fow systems. The main aim of this paper, given the signifcant recent developments in experimental techniques and the availability of solid materials and fuid substances, is to act as an updated, comprehensive resource that consolidates important optical data along with other relevant information on a large number of suitable solid and fuid options currently available for selection, and to provide broader guidance to experimentalists performing detailed, high-fdelity, accurate RIM optical-based measurements in the aforementioned fuid-fow systems. Information is included on aspects such as safety, toxicity, material compatibility, the role of temperature, solubility/ miscibility, and more, as well as the use of liquid mixtures and additives (e.g., salts) for the tuning of properties such as density, viscosity, and surface/interfacial tension.

Multiphase interfacial flows consist of two or more immiscible phases, wherein a phase can be either a gas, liquid, or solid. Multiphase fows are of crucial importance in many diverse settings and applications across a wide range of scales including, but not limited to, fows in biological

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and biomedical systems, atmospheric and marine environments, geological processes such as volcanic flows and landslides, planetary atmospherics, as well as processes in the oil and gas, petrochemical, energy, nuclear, transport, automotive, manufacturing, and food production industries. In particular, multiphase flows are encountered in important processes such as evaporation, condensation, boiling, mixing, reaction, cavitation, erosion, sedimentation, and extraction. Liquid–liquid fows are multiphase fows that consist of only two immiscible liquid, e.g., aqueous and organic, phases. The liquid phases can be present in these flows in a number of different so-called regimes, e.g., (i) stratified or stratified-wavy flows possibly with droplets of either or both phases at or near the interface, which are often encountered in oil and gas transportation lines; (ii) dispersed emulsions, which are common in dairy and chocolate processing, and power generation engines and plants; and (iii) combined emulsions and suspensions, e.g., in paint production. Despite their ubiquitous and important practical relevance, these fows are inherently complex, nonlinear, and multiscale in nature and remain poorly understood.

Optical diagnostic techniques are some of the most established fow-measurement methods. The collection of reliable quantitative data, however, has been dependent on the development of systems necessary for the rapid illumination and associated detection or imaging of interrogated fow regions, including lasers, cameras, photodetectors, etc., as well as the processing capabilities to perform semi-automatic processing of the resulting data. These capabilities only started to become available in the 1970s, which has led to a rapid increase in the evolution and use of these techniques. In particular, state-of-the-art experimental studies of multiphase flows can be performed using advanced non-intrusive, spatiotemporally resolved optical techniques, such as:

- (i) Laser-induced fuorescence (LIF), as described by Kinsey [\(1977](#page-34-0)), Liu et al. ([1977\)](#page-35-0), Walker ([1987\)](#page-37-0) and Crimaldi ([2008\)](#page-33-0), has been used to provide phenomenological insight as well as quantitative information on the distribution and other key characteristics of the fuid phases, including phase fraction, wave amplitude and frequency, bubble size distribution and frequency, etc. LIF has been utilized in RIM systems by Diez et al. [\(2005](#page-33-1)), Ovdat and Berkowitz [\(2006](#page-36-0)), Liu et al. [\(2006a,](#page-35-1) [b\)](#page-35-2), Ravelet et al. ([2007\)](#page-36-1), Wu et al. [\(2011](#page-37-1)), Zadrazil et al. [\(2014](#page-38-0)), Zadrazil and Markides [\(2014](#page-38-1)), amongst others.
- (ii) Laser Doppler velocimetry (LDV), laser/phase Doppler anemometry (LDA/PDA), and similar approaches have been detailed by Durst et al. [\(1976](#page-33-2), [1997](#page-33-3)), Buchhave et al. [\(1979](#page-32-0)), Tropea [\(1995](#page-37-2)), Albrecht et al. [\(2003](#page-32-1)) and Czarske [\(2006](#page-33-4)). These methods allow the measurement of local velocity and/or droplet size

within an interrogated volume and have been used in RIM systems by Varty [\(1984](#page-37-3)), Yarlagadda and Yoganathan [\(1989](#page-38-2)), Walker et al. [\(1989](#page-37-4)), Liu et al. [\(1989](#page-35-3)), Duncan et al. [\(1990](#page-33-5)), Chen and Kadambi [\(1990](#page-33-6)), Wildman et al. [\(1992](#page-37-5)), Koh et al. [\(1994](#page-34-1)), Jana [\(1995](#page-34-2)), Perktold et al. ([1997\)](#page-36-2), Dietze et al. [\(2009](#page-33-7)), and others.

(iii) Particle image/tracking velocimetry (PIV/PTV) and variants thereof have been covered in detail by Adrian [\(1986](#page-31-0), [1991\)](#page-31-1), Arroyo and Greated [\(1991](#page-32-2)), Maas et al. [\(1993](#page-35-4)), Grant ([1997\)](#page-34-3) and Fu et al. ([2015\)](#page-33-8), among others. These techniques can provide velocity information in two or three dimensions (2-D/3-D) and have been used in many RIM systems, e.g., by Northrup et al. ([1991\)](#page-36-3), Peurrung et al. [\(1995](#page-36-4)), Zachos et al. [\(1996](#page-38-3)), Hopkins et al. ([2000\)](#page-34-4), Longmire et al. [\(2001](#page-35-5)), Bale-Glickman et al. [\(2003](#page-32-3)), Ninomiya and Yasuda [\(2006](#page-36-5)), Burgmann et al. ([2009\)](#page-32-4), Dietze et al. ([2009\)](#page-33-7) Buchmann et al. ([2010\)](#page-32-5), Berard et al. ([2013\)](#page-32-6), Im et al. [\(2013](#page-34-5)), Yagi et al. ([2013\)](#page-37-6), Morgan et al. [\(2013](#page-36-6)), Zadrazil and Markides ([2014\)](#page-38-1), Krug et al. [\(2014](#page-35-6)).

In addition, less involved but equally important in providing (semi-)qualitative insight into the fows of interest is direct (high-speed) imaging, based on which a wealth of phenomenological information has been generated allowing signifcant advancements in relevant felds. All the aforementioned techniques have been presented here in the context of multiphase fow measurements; nevertheless, similar issues apply to internal single-phase (liquid) flows, both internal and external when walls are present, since optical distortions can occur at the solid–liquid interfaces (walls), limiting access to near-wall regions.

These optical methods have gained popularity, thanks to several advantages that they offer compared to more classical techniques for multiphase fow measurement and characterization, such as hot-wire/hot-flm anemometry (Ueda and Tanaka [1975](#page-37-7); Majithia et al. [2008](#page-35-7)), parallel/twin-wire, or conductivity probes (Han et al. [2006](#page-34-6); Zhao et al. [2013](#page-38-4)). In particular, optical techniques: (i) can provide both qualitative and detailed high-resolution quantitative information on important fow phenomena and quantities; and (ii) are non-intrusive, so that these flow characteristics can be measured without introducing external disturbances to the fow. The main challenge when implementing for these techniques, however, is the requirement for optically undisturbed or fully known optical paths throughout the interrogated region, covering all fuid phases and any suspensions that may be of interest, including of the illumination and/or the observation or detection (e.g., refected, scattered, and fuorescent) light. Specifcally, optical distortions and intensity variations in the fuid domains under observation can arise due to the refraction and/or refection of light when this passes through materials with differing refractive indices (RIs), i.e., solid–gas, liquid–gas, solid–liquid, and liquid–liquid, depending on the fuids/fows of interest. These distortions lead to unwanted optical warping, displacement, and rotation of the measuring plane or volume, giving rise to increased errors in the measurement of interfacial topologies, velocities, or other scalars. This is typically addressed in the experimental design stage by the minimization of curved, angled, and uneven surfaces with the use of correction boxes similar to that shown in Fig. [1.](#page-2-0) Further improvements can be achieved by placing and then imaging pre-defned graticule targets in the measurement region like that shown in Fig. [2.](#page-2-1) The resulting images of these targets can then be used for spatial corrections during post-processing. Examples of the graticule correction method can be found in Zadrazil et al. [\(2012](#page-38-5)) in single-phase (liquid) circular pipe fows, and Morgan et al. ([2013](#page-36-6), [2016\)](#page-36-7) in two-phase (liquid–liquid) pipe flows. Nevertheless, these corrections and, therefore, the associated errors can be minimized by selecting solid materials and liquid substances that have closely matching RIs, a practice which is known as refractiveindex matching (RIM). The technique of refractive-index matching limits the physical process that causes refraction as well as refection between differing materials.

The refraction of light at an interface is caused by a variation in the optical speed in the two adjacent transmission media, which creates a directional change in the propagation light path. The degree of change of the light path direction



**Fig. 1** Optical correction box illuminated by a laser sheet and containing a test pipe which is both flled and surrounded by an RImatched  $(RI = 1.459)$  fluid. In this case, the pipe material is fused quartz and the oil Exxsol D140

<span id="page-2-0"></span>

<span id="page-2-1"></span>**Fig. 2** Graticule (printed target) for the spatial calibration of images, e.g., within circular pipes

depends on the angle of incidence and the relative difference in the optical speeds in the two transmission media. The speed of light in a transmission medium *v* can be stated relative to the speed of light in a vacuum *c*, through the refractive index (RI), as defned by the following equation:

$$
n = \frac{c}{v},\tag{1}
$$

based on which, Snell's law, also known as is the law of refraction and given in Eq. [2,](#page-2-2) relates a light ray's angle of incidence to its angle of refraction at the interface between two transmission media of differing RIs (here,  $n_1$  and  $n_2$ ; see also Fig. [3](#page-2-3)):

<span id="page-2-2"></span>
$$
\frac{\sin(\theta_1)}{\sin(\theta_2)} = \frac{n_2}{n_1}.\tag{2}
$$

It emerges from Snell's law that larger directional changes (distortions) occur at an interface when: (i) there are greater relative RI mismatches between the two transmission media; and (ii) the incident angle (relative to the interface normal direction) is large, as illustrated in Fig. [3a](#page-2-3)–c. Importantly, this also means that a higher RI mismatch or incident angle can lead to greater measurement errors if not fully corrected. Furthermore, if light travels from a material of higher RI  $(n_1)$  to one of lower RI  $(n_2)$ , then a critical angle  $\theta_c$  exists at which light travels along the interface (Fig. [3](#page-2-3)d), and beyond which the light is fully refected (Fig. [3](#page-2-3)e) creating areas that are optically inaccessible. Snell's law can be used to determine this critical angle  $\theta_c$ , as given by the following equation:

$$
\theta_c = \arcsin\left(\frac{n_2}{n_1}\right). \tag{3}
$$

In summary, the optical illumination of a targeted region of interest or its observation through interfaces (liquid–liquid or solid–liquid) associated with mismatched RIs can introduce positional or intensity uncertainty, and even lead



<span id="page-2-3"></span>**Fig. 3** Refraction of light at an interface as it passes from a high to a low RI medium: *a* no refraction/refection occurs when the light direction is perpendicular to the interface; *b* slight refraction at low angles; *c* increasing refraction at higher angles; *d* critical angle between refraction and refection; and *e* total internal refection

to the observation of multiple images or optically inaccessible regions, especially if the interfaces are moving or curved. Such problems were reported, e.g., by Lowe and Kutt [\(1992](#page-35-8)), who were motivated to develop a spatial correction method using images from two cameras and ray tracing equations.

It should be noted that the RI and thus the angle of refraction are wavelength dependent, and this can lead to varying degrees of optical distortion depending on the wavelength if monochromatic light is used, or dispersion if mixed wavelength light is used; in fact, this forms the basis of some measurement methods. For example, in the case of the most common liquid, pure water, the RI shows rational function dependency on the wavelength in the visible part of the electromagnetic spectrum, with values in the range 1.330–1.343 for wavelengths 405–707 nm at 20 °C/1 bar(a) (Thormahlen et al. [1985\)](#page-37-8). By convention, reported RIs are often measured using the sodium D-line at 598 nm; however, common lasers employed in optical-based measurements emit at different wavelengths (e.g.,  $\lambda_{Nd:YAG} = 266$ , 532, 1064 nm;  $\lambda_{Nd:YLF} = 527$  nm). This can induce errors in RI matching, since the RI of a solid phase or a liquid phase generally decreases with increasing wavelength (Forziati [1950\)](#page-33-9), even in RIM systems that have been matched at their literature-reported values. Patil and Liburdy ([2012\)](#page-36-8) found that the use of fuorescent-dye doped seeding-particles introduced a further source of error, because the emitted fuorescent-light wavelengths differed to that of the laser source, and for which the system was optimized.

Beyond the illumination and/or observation wavelengths, a number of experimental parameters can lead to changes in the RIs of individual materials/substances in optical fuid-fow measurements, thus increasing errors even for initially matched systems in quiescent and laboratory-controlled environments, in particular, variations in temperature or pressure. Mondy et al. [\(1986](#page-36-9)) reported that the temperature had to be kept within  $\pm 1$  °C to observe through 0.15 m of a 30% concentrated solid suspension, due to the temperature dependence of the RI of the fuid. This is an important consideration given that, unless controlled, the temperature can easily vary during experiments due to changes to the ambient conditions or the presence of heat sources or sinks in the fow loop (e.g., pumps or uninsulated walls). Signifcant temperature gradients leading to RI variations that introduce measurement errors were mentioned by Schmidt et al. [\(1984](#page-37-9)) who needed to create a laser-path correction curve to reduce positional errors in their LDA measurements in a fuid with a large temperature gradient. RI deviations can also occur through processes such as hygro-scopicity or evaporation causing relative changes to the components within fuid mixtures. In one example, Miller et al. ([2006\)](#page-35-9) found that the RI of their DEP and ethanol mixture changed over time due to the differential evaporation of the ethanol, highlighting the need for closed systems and fuid loops especially when dealing with volatile fuids. Similarly, changes can occur when the solid materials can absorb the liquid phase(s), leading to RI changes over time (Dijksman et al. [2012](#page-33-10)), or due to the chemical instability or reactivity of the materials employed, again, unless these are controlled by the experimental design and procedure. For instance, Averbakh et al. ([1997\)](#page-32-7) and Shauly et al. [\(1997](#page-37-10)) utilized a mixture of 14.1 wt% 1,1,2,2-tetrabromoethane, 35.7 wt% polyalkylene glycol oil, 50.3 wt% Triton X100, and 0.1 wt% Tinuvin 328 to match the RI of PMMA. In this example, the Tinuvin 328 was specifcally added to the mixture to reduce the rate of UV breakdown of tetrabromoethane. Finally, the RIs of solid components can also vary due to non-uniformities introduced during their manufacture, such as material impurities, inclusions, etc.

Over and above the issues above relating directly to the differences in the RIs, the size of the measurement errors due to RI mismatches depends on many factors including the number of interfaces to the point of measurement and whether these are moving, the quantities being measured, the selected technique, the characteristics of the optical confguration and of the equipment used, and the experimental procedure and post-processing methods. The measurement error for a given fuid RI mismatch can, therefore, only be determined once all these factors have been taken into consideration. Consider, for example, two different types of flow of interest with identical optical setups and methods; in stratified liquid–liquid flows with a single, continuous, and stable interface, large RI differences can be tolerated for high-accuracy measurements, whereas in a finely dispersed liquid–liquid flow or a concentrated solid– liquid suspension, much closer RI matching would be required to achieve similar accuracies for the same measured quantities.

Nevertheless, particle position and consequent PIV measurement errors caused by mismatched RIs were considered by Patil and Liburdy [\(2010](#page-36-10), [2012](#page-36-8)), who measured optical distortions in fows through packed beds and through porous media with RI differences as low as 0.0005. Two main types of distortion were identifed in the former: (i) image centroid distortion due to refraction; and (ii) image intensity distortion, while in porous media, RI mismatching was found to give rise to the formation of multiple particle images, and relative PIV errors amounting to 2 and 4% for absolute RI differences of 0.0016 and 0.0036, respectively. Dijksman et al. [\(2012](#page-33-10)) found, experimentally, that imaging through 15 layers of 3-mm glass spheres was limited to a maximum RI mismatch of about 0.003 due to light scatter, and performed a numerical ray tracing exercise to quantify the effect of RI mismatching between the liquid and solid particles, fnding that a standard deviation of 0.001 in the

spread of the particle RI created the same level of image blur as a RI mismatch of 0.002. Ray tracing also showed that considerable blurring was present through 25 layers when the RI was mismatched by 0.002. The same authors also noted that the RIs of particles are not normally given to the 0.2% accuracy required for RIM, and, therefore, that in situ RI matching was needed in their case. Furthermore, according to Hannoun [\(1985](#page-34-7)), RI differences as small as 0.0001 can affect LDV measurements through 30 cm of RI fuctuations, while Daviero et al. [\(2001\)](#page-33-11) calculated that differences as low as 0.00005 may be needed for 40 cm of RI fluctuations. Hirsch et al.  $(2015)$  $(2015)$  $(2015)$  found that surface roughness, scratches, inclusions, fractures, inhomogeneity, and other manufacturing effects in plastics and glasses can all lead to measurement errors even for apparent perpendicular surfaces; some of these effects were directional causing the magnitude of the error to be depending on the surface axis.

Important parameters beyond optical clarity and accuracy also need to be considered when selecting liquids and solids for a given experiment, including densities and viscosities, non-Newtonian rheological behaviour where applicable, interfacial tension, and surface wettability, which is dependent on the surface energy balance between all fuid and solid phases, and quantifed by the contact angle. Figure [4](#page-4-0) demonstrates the defnition of the contact angle for two fuid droplets on a solid surface in an immersion fuid. The fuid of droplet (a) on the left is more phobic of the surface than the immersion fuid and, therefore, has a larger contact angle, while the fuid of droplet (b) on the right has a greater affnity for the surface than the immersion fuid and so has a smaller contact angle. In particular, surface and interfacial tension should be taken into account when selecting fluids for liquid–liquid flow studies to match the experimental conditions in a given application (e.g., fow of water and oil through crude-oil transport pipelines).

Moreover, low reactivity and mutual solubility are desired, the latter being important in preventing the liquid–liquid interface becoming indeterminate. Similarly, the presence of surfactants in either fuid phase, whether



<span id="page-4-0"></span>**Fig. 4** Two droplets on a solid surface in a lighter emersion fuid (*gas* or *liquid*) making contact angles *θ* with a solid. The *liquid* of droplet (**a**) has a lower affnity for the surface than the immersion fuid and, therefore, has a large contact angle, while the *liquid* of droplet (**b**) has a greater affnity for the solid than the immersion fuid and so has a smaller contact angle

intentionally or not, can lead to the formation of emulsions and cause surface effects such as varying surface and interfacial tension across position and time. Finally, stability, toxicity, fammability, compatibility, and laser power tolerance should all be considered and carefully controlled. The design guidelines for liquid–liquid fow systems of Smedley and Coles ([1990\)](#page-37-11) are particularly useful here. Generalizing these guidelines, it is suggested: (i) not to use any system components, liquids or solids, that are unstable, reactive either between themselves or with the environment (air, fow loop components, etc.); (ii) not to use liquids that either damage, absorb, or dissolve any experimental solids or components of solids like plasticizers; (iii) not to use chemicals or optical components that undergo photodissociation or damage especially at wavelengths and powers used in light sources/lasers; (iv) to use safe, low-toxicity, and low-fammability chemicals whenever practicably possible; (v) to use materials that are optically transparent at all experimental wavelengths; (vi) to avoid liquids with high vapour-pressures or that are hydroscopic, thus reducing RI changes due to evaporation or absorption; (vii) to select low-cost fuid options; (viii) to select liquids that match the RI of the observations solids (walls, particles); and (ix) to select solids and liquids that give any desired contact angles. In the case of liquid–liquid systems, specifcally, select the components for each phase to: (i) have low interphase solubility; (ii) allow matching over the required range of viscosities and densities; (iii) allow matching over a wide range of RIs to extend the options for the observational solid; and (iv) provide the desired interfacial tension between the liquids.

Four comprehensive papers have been published on RIM systems (Smedley and Coles [1990;](#page-37-11) Budwig [1994;](#page-32-8) Wiederseiner et al. [2011;](#page-37-12) Dijksman et al. [2012](#page-33-10)). Smedley and Coles ([1990](#page-37-11)) created 121 single-component (pure) immiscible liquid–liquid pairs, some of which had closely matched RIs; however, none of these RIM pairs were based on the common fuids of water, glycerol, or silicone oil, and were not RI tuned as both liquids were single-component. Bud-wig [\(1994\)](#page-32-8) detailed the principles of RIM liquid flows covering both single-phase liquid and two-phase liquid–liquid flows. Wiederseiner et al.  $(2011)$  $(2011)$  reviewed the RIM literature for flows containing concentrated particle suspensions and covered solid–liquid systems in great detail, including techniques for matching the RI of solid particles and liquids. Dijksman et al. [\(2012\)](#page-33-10) reviewed dense granular RIM systems for use with 3-D tomographic techniques whilst detailing the optical effect of RI mismatches in such systems. The tomographic method relied on laser scanning to build 3-D data of the granular material through the RIM system. Importantly, to the best of our knowledge, no publication to date has focused specifcally on and documented earlier liquid–liquid and multiphase solid–liquid fow systems.



**Fig. 5** Raw image from laser illumination in a stratifed-wavy Exxsol D140 oil and water fow, showing planar LIF (PLIF) from the added dye in the water phase at the *bottom* of the pipe and particles for PIV/ PTV in both *liquid* phases

<span id="page-5-0"></span>By means of clarifcation, in some liquid–liquid RIM systems, only one of the liquids is matched to the solid, while the other is unmatched (e.g., due to other experiment design restrictions). Figure [5](#page-5-0) shows such a system with Exxsol D140 oil and water. The oil is matched to the quartz pipe section at a RI of 1.459, whilst the water is unmatched with a RI of 1.333, resulting in distortions at the interface and in the water layer when viewing the flow from the bottom of the pipe; see the ghost-like refections that are present around the liquid–liquid interface in the figure. For the purposes of this review, this type of liquid–liquid system is treated as if it is a solid–liquid RIM system (Sect. [2](#page-5-1)), and only systems where both liquids are matched are treated as liquid–liquid RIM systems (Sect. [3](#page-16-0)). These liquid–liquids RIM systems are then optionally matched to a solid material (Sect. [3.2\)](#page-22-0).

The paper is organized as follows: Sect. [2](#page-5-1) reviews solid–liquid RIM systems; Sect. [3](#page-16-0) reviews liquid–liquid and solid–liquid–liquid RIM systems; Sect. [4](#page-22-1) discusses liquid options and further RI tuning. Finally, conclusions are drawn in Sect. [5.](#page-30-0)

### <span id="page-5-1"></span>**2 Solid–liquid systems**

This section reviews solid–liquid fow systems that have previously been utilized in RIM experiments, i.e., matching the RI of a particular solid with either a pure liquid, a mixture of liquids, or additive solutions. Solid–liquid systems can be understood here as a suspension of particles in a liquid continuum (e.g., fow of a sand slurry) or a single-phase flow (e.g., external flow over a solid obstacle or internal flow within a pipe or conduit). An example of a solid–liquid system is given in Fig. [6,](#page-5-2) and consists of a 32-mm fused quartz pipe inside a correction box, where the box and pipe are both filled by a RIM Exxsol D140 oil ( $RI = 1.459$ ).

A large number of solid–liquid RIM systems have been previously employed and so for clarity, these are separated into the following four sub-sections: Sect. [2.1](#page-5-3) deals with common plastics; Sect. [2.2](#page-11-0) considers silicone and urethane



<span id="page-5-2"></span>**Fig. 6** Fused quartz pipe located inside a correction box with RIM liquid Exxsol D140 (RI = 1.459) both surrounding the *box* and within the pipe

rubbers; Sect. [2.3](#page-12-0) reports on custom polymers, resins, and hydrogels; and fnally, Sect. [2.4](#page-15-0) focuses on glasses.

#### <span id="page-5-3"></span>**2.1 Common plastics**

Plastics are a common material for conducting RIM experiments. Many plastics are transparent or translucent when in the amorphous state, while the transparency of plastics in the crystalline state can sometimes be improved through transitioning to a more transparent amorphous state through heating above the melting temperature and quenching (Wiederseiner et al. [2011\)](#page-37-12). Plastics are also readily available, affordable and can be easily manufactured in many different shapes and sizes. Consequently, they are preferentially chosen for the study of fows in complex geometries, e.g., biological systems, moving machinery, etc. (Soranna et al. [2008](#page-37-13); Bale-Glickman et al. [2003](#page-32-3)). Two manufacturing processes are commonly used for the production of plastic components (e.g., test sections) that are commonly employed in scientifc studies, namely extrusion and casting; more recently, rapid prototyping and 3-D printing processes have allowed an even greater range of components to be made from suitable plastics. Depending on the process, plastic components often bear different physical properties, despite their chemical composition being the same. Plastic pipes are frequently produced via extrusion processes, which can result in a ribbed effect on the pipe walls and a variability (and asymmetry) in the pipe diameter. Nevertheless, plastic pipes typically have a roughness of  $\sim$ 1  $\mu$ m, which is much smoother than steel pipes frequently installed in industrial (e.g., the oil-and-gas) systems that have a roughness of 10s of  $\mu$ m (Hydraulic-Institute [1979](#page-34-9)).

Plastics vary widely in chemical composition and, as a result, important properties like air–water–solid contact angles  $\theta$  (see Fig. [4](#page-4-0)) can span a wide range of values. For example, Nylon 6 has  $\theta = 70^{\circ}$  (Fort [1964](#page-33-12)), while the highly hydrophobic fuorocarbon polytetrafuoroethylene (PTFE) has  $\theta = 118^\circ$  (Zhang et al. [2004](#page-38-6)). Plastics also vary in chemical resistance, so compatibility with contacting fuids should be checked carefully, since any incompatibility can lead to damage to the test section or other components in the flow loop. Dijksman et al. [\(2012\)](#page-33-10) found that poly(methyl methacrylate) (PMMA) can develop microcracks when washed and dried after being in contact with Triton X100. Nevertheless, the chemical resistance of plastics can, in some cases, be improved. In the case of microcracks in PMMA, this can be done by reducing internal stresses through annealing after cutting (Hendriks and Aviram [1982\)](#page-34-10). Further chemical compatibility issues encountered in the literature are discussed in the corresponding material section. Apart from chemical damage, physical damage can also occur from lasers, and this can occur at relatively low laser powers in some plastics, such as polycarbonate and PMMA [e.g., compared to glass, Hirsch et al. ([2015](#page-34-8))]. While this is mainly a consideration when using high-powered lasers, it should still be reviewed for a given experiment, especially if high luminosities are required.

This section focuses on solid–liquid matched RI systems, where the solid phase consists of a plastic material. The following plastics are considered, in order of increasing RI: fuorinated ethylene propylene (FEP), poly(vinyl acetate) (PVA), PMMA, nylon, and polystyrene (PS). In addition, previously unused, but common, plastics that could be utilized in future RIM experiments are included, namely: tetrafuoroethylene–hexafuoropropylene–vinylidene fuoride (THV), ethylene–tetrafuoroethylene (ETFE) and polyvinylidene difuoride (PVDF). Collated data of a number of possible solid–liquid RIM combinations, together with the known density and viscosity values of pure or fuid mixtures, can be found in Table [1.](#page-7-0)

Referring to Table [1](#page-7-0), we proceed here to provide details relating to the various solid plastic materials, along with dedicated examples of their use in RIM systems. FEP is a hydrophobic fuorpolymer with a RI of 1.338, a density of 2150 kg/m<sup>3</sup>, an air-water contact angle of  $\theta = 102.1^{\circ}$ (Akinci and Cobanoglu [2009\)](#page-31-2), and has good chemical resistance. At optical wavelengths, FEP is translucent rather than clear, yet despite this, good images can still be taken through FEP walls a few mm thick. Importantly, the RI of FEP is close to that of water (1.333), and as a result, FEP has been implemented extensively as a RIM solid with water. For example, Satake et al. [\(2015](#page-37-14)) performed holographic PTV measurements in a sphere-packed pipe using a RIM system with water as the test fuid, an FEP pipe  $(RI = 1.338)$ , and spheres were made from MEXFLON resin ( $RI = 1.330$ ). The pipe was located in a square observation section flled with water (correction box) to reduce distortions from the outside pipe wall.

PVA is a rubbery polymer with a RI of about 1.470, a density of 1190 kg/m<sup>3</sup>, and an air-water contact angle of  $\theta$  = 60.6° (McCafferty and Wightman [1999\)](#page-35-10). A fluid mixture of Pale 4 oil (oxidized castor oil) and tetrabromoethane was created by Karnis et al. [\(1966](#page-34-11)) to match the RI of PVA discs with  $RI = 1.467$ . This RIM system was used for the optical study of dilute, disc-shaped particle suspensions in Couette and Poiseuille fows. The authors reported satisfactory results with PVA in their application.

PMMA, which is also known by the trade names Perspex, Plexiglas, or acrylic glass, is the most commonly employed solid in RIM experiments with a RI of  $\approx 1.490$ , a density of  $1180 \text{ kg/m}^3$ , and an air-water contact angle of  $\theta$  = 59.3° (Stöhr et al. [2003\)](#page-37-15). It is optically clear and is easily machined, although it is also brittle and prone to cracking. It has been used for optical measurements in RIM systems with complex geometries, e.g.: a model of abdominal aortic aneurysms (Budwig et al. [1993;](#page-32-9) Egelhoff et al. [1999](#page-33-13)); a column containing dispersed beads (Haam and Brodkey [2000;](#page-34-12) Haam et al. [2000\)](#page-34-13); an axial turbo-pump (Uzol et al. [2002,](#page-37-16) [2007](#page-37-17)); or highly concentrated (50%) spherical suspensions (Breedveld et al. [1998](#page-32-10), [2001,](#page-32-11) [2002;](#page-32-12) Breedveld [2000\)](#page-32-13). PMMA can, however, be attacked by a number of chemicals, and hence, liquid compatibility must be carefully considered. Chemicals including ethanol (and other alcohols), dimethylsulfoxide (DMSO), and Triton X100 can all cause PMMA to form microcracks (Dijksman et al. [2012](#page-33-10)). In a related study, Liu et al. ([1990\)](#page-35-11) found that stress cracks and crazing created in PMMA by a mixture of 68.2 vol% turpentine oil and 31.8 vol% Tetralin ( $RI = 1.489$ ) could be removed by a careful casting, machining and subsequent annealing treatment. Similarly, Jana [\(1995](#page-34-2)) treated PMMA with a silicon-based hard coating to increase its chemical resistance to Triton X100, which despite causing crazing is a close RI match to PMMA. Figure [7b](#page-11-1) shows a rod of PMMA immersed in Triton X100 with an RI of 1.489, and for contrast, Fig. [7](#page-11-1)a shows the same PMMA rod in air. Terpineol also attacks PMMA over long time scales, as observed Mondy et al. [\(1986](#page-36-9)) who matched PMMA to a mixture of 33.7 wt% polyalkylene glycol oil, 41.8 wt% terpineol, 24.4 wt% 1,1,2,2-tetrabromoethane, and 0.1 wt% Tinuvin. Furthermore, Dijksman et al. ([2012](#page-33-10)) noted that PMMA is known to absorb water and found that it will also absorb Triton X100 as well as dimethylsulfoxide, and that this absorption leads to changes in the RI of PMMA, while Lyon and Leal ([1998a\)](#page-35-12) found that the RI of their quaternary liquid mixture was affected due to the differential absorption of the 1,6-dibromohexane mixture component into PMMA particles. Numerous liquids have been utilized for the matching of PMMA's RI, including aqueous salt solutions, and binary and ternary mixtures. A list of PMMA RIM systems along with density and viscosity information can be found in Table [1.](#page-7-0)

Nylon is translucent plastic, like FEP, with an RI of about 1.510, a density of about 1150 kg/ $m<sup>3</sup>$  and has an

<span id="page-7-0"></span>



**Table 1** continued

Table 1 continued



**Table 1** continued



 $\overline{\phantom{a}}$ 

**Table 1** continued

Table 1 continued

 $\overline{\phantom{a}}$ 



<span id="page-11-1"></span>**Fig. 7** Photographs of a PMMA rod in: **a** air, demonstrating large optical distortions; and **b** RIM Triton X100 liquid (RI = 1.489)

air–water contact angle of  $\theta = 70^{\circ}$  (Fort [1964\)](#page-33-12). Our literature search has shown that Nylon has rarely been used in RIM experiments. Nevertheless, Nylon rods with a  $RI = 1.514$  have been matched to a blend of Pale 4 oil (oxidized Castor oil) and tetrabromoethane to study optically dilute rod-shaped particle suspensions in Couette and Poiseuille fows (Karnis et al. [1966\)](#page-34-11).

Polycarbonate (PC) is another common clear hard plastic with a RI of  $\approx 1.580$ , a density of approximately 1210 kg/m<sup>3</sup>, and an air-water contact angle of  $\theta = 78^\circ$ (Cho et al. [2003\)](#page-33-24). Its relatively high RI means that combinations with aqueous solutions are rare, and as a result, no PC RIM experimental systems were found in the literature. Hendriks and Aviram ([1982\)](#page-34-10), however, showed that a zinc iodide  $(ZnI<sub>2</sub>)$  solution could be created to match PC. Even so, Hirsch et al. [\(2015\)](#page-34-8) found that distortions through polycarbonate windows lead to lower accuracy position measurements than with PMMA or glass, and that polycarbonate was also damaged by lower laser powers than PMMA and glass. As a result of these fndings, Hirsch et al. [\(2015\)](#page-34-8) recommended that polycarbonate should not be used for optical measurements.

Finally, referring to Table [1,](#page-7-0) PS is a transparent plastic with an RI between 1.590 and 1.600, a density of about 1050 kg/m<sup>3</sup> and an air-water contact angle of  $\theta = 90^\circ$ (Kondyurin et al. [2006\)](#page-34-28). Of note is the fact that PS is vulnerable to a number of chemicals, such as cyclohexyl bromide and decalin (Dibble et al. [2006\)](#page-33-21), and its use as a RIM solid in the literature is rare. However, one application example by Koh et al. [\(1994\)](#page-34-1) had particles made from PS with divinylbenzene cross-linkage that were both density and RI (1.600) matched to a mixture of 1-methylnaphthalene, 1-chloronaphthalene, and polyalkylene glycol. It was suspected, however, that the PS particles slowly absorbed both the 1-methylnaphthalene and 1-chloronaphthalene causing the RIs of both the solids and liquids to vary over time (Koh et al. [1994](#page-34-1)). The relatively high RI of PS generally restricts the available fuid RIM options. Although no matching aqueous systems found in the literature,

Hendriks and Aviram  $(1982)$  showed that a ZnI<sub>2</sub> aqueous solution can match PS.

A number of additional commercially available transparent or translucent solids of interest can be found on the market that have not yet been employed in RIM experiments to the best of the authors' knowledge. These include THV, ETFE, and PVDF, which are all hydrophobic fuoropolymers with RIs of 1.350, 1.403, and 1.420, densities of 1970, 1730, and 1780 kg/m<sup>3</sup>, and air-water contact angles of  $\theta = 99^{\circ}$  (Begolo et al. [2011](#page-32-23)),  $\theta = 99.2^{\circ}$  (Akinci and Cobanoglu [2009\)](#page-31-2), and  $\theta = 94^{\circ}$  (Saarinen et al. [2006](#page-36-19)), respectively. ETFE and PVDF are optically translucent rather than clear in the visible part of the electromagnetic spectrum (although slightly less clear than FEP), whereas THV is transparent. Both ETFE and PVDF are relatively stiff fuoropolymers allowing thin-walled tubes and other sections with adequate optical transparency to be constructed. As an example, the optical clarity of a 50-mm ID ETFE pipe with a 1-mm-thick wall can be seen in Fig. [8,](#page-12-1) while the optical clarity of a 14-mm ID PVDF pipe with a 0.7-mm-thick wall can be seen in Fig. [9](#page-12-2). Similar to other fuoropolymers, like FEP, these materials have excellent chemical resistance. Of course, even with the mention of these plastics, this is not a complete list, especially since new materials are constantly emerging. Further plastic RIM options have been suggested by Wiederseiner et al. [\(2011](#page-37-12)), including polyetheretherketone (PEEK), polyethylene terephthalate (PET), polyvinylchloride (PVC), styrene/acrylonitrile copolymer (SAN), polychlorotrifuoroethylene (PCTFE), and polyformaldehyde (POM), with RIs of 1.650–1.770, 1.575, 1.540, 1.570, 1.435 and 1.410, respectively.

### <span id="page-11-0"></span>**2.2 Silicone and urethane rubbers**

Silicone rubbers are elastomers composed of a siliconcontaining polymer. They typically have RIs in the range 1.410–1.440, while the air–water contact angle of PDMS silicone rubber is 108° (Duffy et al. [1998](#page-33-25)). The density of



**Fig. 8** Photographs of an ETFE pipe with a 50-mm ID and 1-mm wall thickness: **a** located within a PMMA fange; and **b** with an internal measurement tape

<span id="page-12-1"></span>

<span id="page-12-2"></span>**Fig. 9** Photograph of a PVDF pipe with a 14-mm ID and 0.7-mm wall thickness with an internal measurement tape

silicone rubber varies widely based on its exact composition; however, it is usually in the range of 1100–2300 kg/ m<sup>3</sup>. Flexibility makes silicone rubbers particularly useful for compliant models of fows through fexible structures or membrane-like tissues, e.g., in RIM models for bloodflow experiments and, as a result, have been frequently employed in such systems (Duncan et al. [1990](#page-33-5); Perktold et al. [1997](#page-36-2); Bale-Glickman et al. [2003;](#page-32-3) Burgmann et al. [2009](#page-32-4); Shuib et al. [2010;](#page-37-25) Yousif et al. [2010;](#page-38-11) Gülan et al. [2012](#page-34-29); Pielhop et al. [2012](#page-36-20); Geoghegan et al. [2012;](#page-33-26) Im et al. [2013](#page-34-5); Kefayati and Poepping [2013\)](#page-34-30). Sylgard 184, manufactured by Dow Corning, has been identifed as a silicone rubber of particularly interest (Duncan et al. [1990;](#page-33-5) Perktold et al. [1997;](#page-36-2) Hopkins et al. [2000](#page-34-4); Yousif et al. [2010](#page-38-11); Shuib et al. [2010](#page-37-25); Buchmann et al. [2010,](#page-32-5) [2011](#page-32-24); Geoghegan et al. [2012](#page-33-26) and Kefayati and Poepping [2013](#page-34-30)). Although a common choice, Hopkins et al. ([2000](#page-34-4)) cautioned that the effects of mixing and curing on Sylgard 184 can result in RI variations between models, and the care must, therefore, be taken in matching liquid RIs to individual models. It should be noted that silicone rubbers are known to absorb some liquids, including silicone oils and methlcyclohexane, and this can lead to signifcant swelling of the material (100% or more) while also potentially affecting the RI (Burdett et al. [1981\)](#page-32-25). This is important in the practical deployment of these materials, as silicone oils are a close RI match for silicone rubbers but are unlikely to be suitable in most cases. An example of the use of silicone rubber is given in Im et al. [\(2013](#page-34-5)), where silicone rubber

was RI matched to a glycerol solution to perform tomographic PIV measurements through a model of a nasal cavity.

Although rarely used, urethane is another transparent rubber that can be employed to study flows in compliant geometries. Le et al. [\(2013](#page-35-18)) matched urethane rubber with a RI of 1.490 to an aqueous solution of sodium iodide (NaI) and glycerol to study a model of an aneurysm. The different liquids identifed as having been used in RIM experiments featuring rubbers are shown in Table [2](#page-13-0).

### <span id="page-12-0"></span>**2.3 Custom polymers, resins, and hydrogels**

Optically clear resins have been developed with a very wide range of customized RIs (covering the range 1.31– 1.60), and are currently available commercially from companies such as MY Polymers and Addison Clear Wave Inc. There are over 50 different types of optically clear resins on the market with varying composition, compatibility, and, of course, RI. Resins are frequently moulded into complex geometries, and can be found in optical equipment including lenses, optical adhesives, and fbre optics. The main advantage of using optically clear resins is that their RIs can be carefully tuned in a similar way to that of a mixture of liquids; however, the cost of these custom-made resins in relatively high may prove prohibitive, especially for large-scale experiments. In one successful implementation, Butscher et al. ([2012\)](#page-32-26) performed PIV measurements in a foam-like porous test section that was entirely made of the epoxy resin WaterShed XC 11122, which was RI matched to the liquid anisole at a RI of 1.515. In another example, Leis et al. [\(2005](#page-35-19)) examined the fuoropolymer–copolymer Nifon as RIM combination with water for studying bioflms. Importantly, Nifon is an atypical hydrophilic fuoropolymer with  $RI = 1.336 - 1.343$  when water wetted.

Hydrogels comprise networks of polymer chains that contain large amounts of water (>90%), leading typically to RIs in the range 1.333–1.349. They have been used as



<span id="page-13-0"></span> $\underline{\textcircled{\tiny 2}}$  Springer

Table 2 List of rubber-based RIM systems

\* Temperature for viscosity is provided when stated; otherwise, it is assumed that this is at ambient laboratory conditions between 20 and 25 °C

\* Temperature for viscosity is provided when stated; otherwise, it is assumed that this is at ambient laboratory conditions between 20 and 25 °C



<span id="page-14-0"></span> $\underline{\textcircled{\tiny 2}}$  Springer

particles in fows but are less likely to be suitable for use as rigid walls as they possess a degree of fexibility. The most common hydrogels include those based on polyacrylamide or agarose (Byron and Variano [2013\)](#page-32-27). Weitzman et al. [\(2014](#page-37-27)) explored the use of copolymers of polyacrylamide and sodium acrylate in creating hydrogels that were RI matched to water while being readily available at low cost and easily moulded. In another successful application, hydrogel spheres and water were also RI matched by Kang et al. [\(2010](#page-34-31)) to perform visualization experiments showing the invasion drainage of porous media with density-matched immiscible liquids. The second liquid phase in these tests, an organic phase consisting of a mixture of soybean oil and carbon tetrachloride, was intentionally unmatched in RI, so that it could be easily identifed and observed. The custom polymers, resins, and hydrogels identifed in the literature as having been used in RIM experiments are listed in Table [3.](#page-14-0)

### <span id="page-15-0"></span>**2.4 Glasses**

Many glasses are readily available with a wide range of RIs (1.45–1.51). Glasses are of interest for RIM experiments as they are optically transparent and provide good chemical resistance, making them compatible with a variety of aqueous and organic fuids. Importantly, glasses can tolerate signifcantly higher laser powers than plastics before undergoing damage, which may be an important factor when higher illumination levels required (Hirsch et al. [2015](#page-34-8)), and they can also withstand higher stresses without signifcant deformation (e.g., due to pressure). On the other hand, glasses are not as easily machined and are typically more brittle than plastics. Glasses are typically hydrophilic and tend to be water wetted, e.g., in liquid–liquid water– oil fows. Common (clean) polished quartz and borosilicate glasses have contact angles in the range 10°–20° (Bowman [1998](#page-32-29)). Furthermore, glasses have a surface roughness of typically  $\sim$ 1  $\mu$ m, which is the same as that of plastics pipes but smoother than metal pipes.

The RI of glass components varies slightly depending on their manufacturing process and composition. In the context of RIM experiments, RI variations should be established to ascertain their effect on measurement accuracy. Dijksman et al. [\(2012](#page-33-10)) found that different batches of glass beads had RIs that varied by up to 0.01. Similarly, the type of glass can affect optical accuracy. Hirsch et al. ([2015\)](#page-34-8) found that optical glass was, unsurprisingly, best suited for optical measurements but also that annealed-normal-glass had only slightly worse optical performance. Our RIM literature search revealed three common glass types: fused quartz; borosilicate; and soda-lime glass. RIM systems featuring these glasses can be found in Table [4,](#page-17-0) along with silica gel.

Fused quartz is high-purity silicon oxide  $(SiO<sub>2</sub>)$  in amorphous (i.e., non-crystalline) form with a density of 2203 kg/ m<sup>3</sup>. It has both a lower coefficient of thermal expansion  $(5.5 \times 10^{-7} \text{ K}^{-1})$  and a lower RI  $(1.450-1.460)$  compared to both borosilicate and soda-lime glasses, although its purity results in it being more expensive than these glasses. Fused quartz has featured in numerous RIM systems, including an RIM facility described by Stoots et al. [\(2001](#page-37-28)) for the purposes LDA flow studies around complex geometries for which the fused quartz observational sections of the facility were RI matched at 1.459 to a light mineral oil ("Penreco Drakeol #5"). McIlroy et al. ([2010\)](#page-35-21) then employed this fow system to study the turbulent fow in a gas-cooled reactor part using tomographic PIV.

Silica gel is a form of  $SiO<sub>2</sub>$  which is often in the form of granules or a porous material with a density of approximately 2210 kg/m<sup>3</sup> and an RI of 1.452. Silica gel is highly hydroscopic with a specific surface area of about 800 m<sup>2</sup>/g. Once it becomes saturated with water, it can be regenerated by heating to 120 °C for 1 to 2 h. Silica gel has been used in a few RIM systems including that of Abbas and Crowe [\(1987](#page-31-6)) who used beads of silica gel that were RI matched to a mixture of chloroform and water to perform LDA measurements on homogenous slurry near transitional Reynolds numbers. Chloroform was selected in this study as it was considered reasonably priced, non-toxic, non-fammable, chemically stable, optically clear, and colourless.

Borosilicate glass, also known as Pyrex and Duran glass, is a laboratory glass with an RI in the range 1.470– 1.474, a density of 2230 kg/m<sup>3</sup>, and a thermal expansion coefficient of  $3 \times 10^{-6} \text{ K}^{-1}$  which is generally lower than soda-lime glass but higher than fused quartz. The literature indicates that borosilicate glass is the second most common solid in RIM experiments behind PMMA. The wide use of borosilicate glass in RIM experiments has resulted in the identifcation of many liquid-phase matching substances (aqueous and organic). Similar to PMMA, borosilicate glass has been used for optical measurements in complex geometries, e.g., an aortic model (Walker et al. [1989](#page-37-4)) and fow through a porous medium (Cenedese and Viotti [1996\)](#page-33-28). Reddy et al. ([2013\)](#page-36-21) created a solid–liquid fuidized bed for PIV measurements, where the solid bed was made from borosilicate glass spheres with a RI of 1.470 and the column was made from PMMA. A number of liquids were examined as matches to borosilicate spheres. A 55 wt% NaI solution ( $RI = 1.475$ ) was RI matched but was considered too corrosive. A 42 wt% potassium thiocyanate (KSCN) solution (RI = 1.460) and a 45 wt% ammonium thiocyanate (NH<sub>4</sub>SCN) solution (RI = 1.470) were also considered reasonable RI matches, but were also regarded too toxic and corrosive. Three further mixtures of benzene and turpentine (mixture  $RI = 1.470$ ), turpentine and chloronaphthalene ( $RI = 1.465$ ), and turpentine

and benzyl alcohol  $(RI = 1.470)$  were good RI matches for the borosilicate glass, but attacked the PMMA test section. A mixture of 68 wt% turpentine and 32 wt % tetralin  $(RI = 1.467)$  was found to RI match the borosilicate spheres, while also being compatible with the PMMA column. Finally, a light paraffn oil with an RI of 1.465 was also found to be a good RI match for borosilicate glass, whilst being compatible with the PMMA. This light paraffin oil was then added to adjust the viscosity of the 68 wt% turpentine and 32 wt% tetralin mixture used as the base experimental liquid.

Soda lime is the most common form of glass with a density of approximately  $2520 \text{ kg/m}^3$ . It is more brittle than both borosilicate and fused quartz, but has a lower cost. It is predominantly composed of silicon dioxide  $(SiO<sub>2</sub>)$  along with other oxides including those of sodium  $(Na, O)$ , calcium (CaO), aluminium (Al<sub>2</sub>O<sub>3</sub>), potassium (K<sub>2</sub>O), titanium (TiO<sub>2</sub>), magnesium (MgO), iron (Fe<sub>2</sub>O<sub>3</sub>), sulphur (SO<sub>3</sub>), and other impurities. The exact composition varies with the manufacturing process, which also affects the RI of the material between batches and suppliers. A search of the literature has shown that soda-lime glass typically has RI values between 1.50 and 1.52 due to this compositional variation. The relatively wide range of soda-lime glass RIs means that care must be taken when matching liquid(s) to solid components. Chen and Fan [\(1992](#page-33-29)) used two NaI solutions to match the RI of soda-lime and borosilicate glasses, in their study of 3-D fow structures in a solid–liquid–gas fuidized bed system. The soda-lime glass beads  $(RI = 1.500)$  of the fluidized bed were matched to a 60 wt% NaI solution, while the borosilicate cylindrical walls were enclosed in a box flled with a 55 wt% NaI solution that matched the RI of borosilicate (1.474). It is not clear why different glasses were selected for the beads and cylindrical walls, as the selection of the same glass would have led to a closer RI-matched system.

The lowest RI of the aforementioned glasses is that of fused quartz ( $RI = 1.450$ ); nevertheless, the RI of calcium fluoride  $(CaF_2)$  glass is even lower at 1.434 (Malitson [1963](#page-35-22)). Given this RI, as well as its insolubility in water,  $CaF<sub>2</sub>$  appears as excellent RIM candidate for use in aqueous-based liquid–liquid systems; however, the authors have been unable to find examples of  $CaF<sub>2</sub>$  employed in this way in the literature. The mineral form of  $CaF<sub>2</sub>$ , fluorite, is often fuorescent under ultraviolet light, and as a result, care should be taken when selecting the purity of the  $CaF<sub>2</sub>$ glass as well as the wavelength of any light-source/laser.

### <span id="page-16-0"></span>**3 Multiphase fow systems**

Optimal optical measurements (high-speed photography, LIF, LDV/PDA, and PIV/PTV) in liquid–liquid RIM systems require that the RIs of the two (or more) liquids and of the observational solids are closely matched. Liquid– liquid systems can be either miscible or immiscible. Miscible liquid–liquid RIM systems, which are considered single-phase flows here, are of interest, for instance when studying the mixing or multicomponent, density stratifed flows (Hannoun [1985](#page-34-7); Hannoun et al. [1988](#page-34-32); De Silva and Fernando [1998](#page-33-30); Daviero et al. [2001](#page-33-11)) or gravity-induced flows (McDougall [1979](#page-35-23); Alahyari and Longmire [1994,](#page-31-7) [1997\)](#page-32-30). The miscible liquids can be RI matched and the density can be tuned separately, for example, Alahyari and Longmire [\(1994\)](#page-31-7) used two aqueous solutions (potassium dihydrogen phosphate  $(KH_2PO_4)$  and glycerol) to create a density difference of 4% in the RIM fuids. Similarly, beyond RI matching, it may be desirable to match also the viscosity of the liquids to set the Reynolds number or, alternatively, to introduce controlled viscosity variations that would be exhibited in processes such as the mixing of miscible systems. Nevertheless, these miscible liquid– liquid systems are not discussed any further, and we continue onto immiscible liquid–liquid systems.

Immiscible liquid–liquid systems are of interest in experiments that investigate complex multiphase fows consisting of two or more liquid phases. The majority of the relevant experiments reported in the literature have been performed in an environment where the liquid phases have been RI matched, but the observational solid phase has a different RI. While this is not ideal, in many cases, any curved solid surfaces (e.g., test-section walls) are static, thus making the optical distortions constant, so that they can be later corrected using the techniques described in the introduction. Consider the example of the liquid–liquid RIM system of Morgan et al. ([2012](#page-36-22)), who applied planar LIF (PLIF) to horizontal liquid–liquid flows in a square test section. In this work, Exxsol D80 was RI matched at 1.444 to an 81.7 wt% glycerol solution. In Morgan et al. ([2013,](#page-36-6) [2016](#page-36-7)), PLIF and PIV/PTV measurements were made using the same fuids but in a circular borosilicate glass pipe which had a RI of 1.474. The distortion caused by the mismatched solid RI was corrected for using a combination of a correction box and the graticule correction method.

In what follows, RIM systems are split into liquid–liquid (Sect. [3.1](#page-16-1)) and solid–liquid–liquid systems (Sect. [3.2\)](#page-22-0).

#### <span id="page-16-1"></span>**3.1 Liquid–liquid systems**

Transparent single-component immiscible liquid–liquid pairs, where one liquid was hydrophobic and the other hydrophilic, were experimentally studied by Smedley and Coles [\(1990](#page-37-11)). In all, they collated 121 compatible systems, of which 13 had an RI match of 0.001 or less, and a further 20 had RI differences of 0.005 or less. Given that all 121

<span id="page-17-0"></span>



**Table 4** continued

Table 4 continued



\* Temperature for viscosity is provided when stated; otherwise, it is assumed that this is at ambient laboratory conditions between 20 and 25 °C

\* Temperature for viscosity is provided when stated; otherwise, it is assumed that this is at ambient laboratory conditions between 20 and 25 °C

## $\underline{\textcircled{\tiny 2}}$  Springer

**Table 4** continued

Table 4 continued

### <span id="page-20-0"></span>**Table 5** List of selected liquid–liquid systems



### **Table 5** continued



\* Temperature for viscosity is provided when stated; otherwise, it is assumed that this is at ambient laboratory conditions between 20 and 25 °C

systems were single component and, therefore, not tuned in RI, it seems likely that many of these systems could be signifcantly improved upon through blending or additives (see Sect. [4](#page-22-1)). The 13 closest RIM pairs of Smedley and Coles ([1990\)](#page-37-11) are included in Table [5](#page-20-0). It is worth noting that none of RIM systems in Smedley and Coles ([1990\)](#page-37-11) involve the commonly used RIM fuids of water, glycerol, or silicone oil. These liquids can, however, be found in many other liquid–liquid RIM systems included in Table [5.](#page-20-0) The RIs of the matched liquid–liquid systems span the range from 1.322 to 1.460, with the majority of actual experimental fow systems being near a value of 1.40.

### <span id="page-22-0"></span>**3.2 Solid–liquid–liquid systems**

Our review of the literature has shown that RIM systems have been used predominantly for experiments featuring solid– liquid systems, and that while liquid–liquid RIM systems are common, full three-phase solid–liquid–liquid RIM systems are rare, with only a few cases reported in the literature. In one such effort, Burdett et al. ([1981](#page-32-25)) investigated a solid–liquid–liquid RIM system for studying dispersed fows, holdup, and axial mixing in packed extraction columns. The matching of liquid–liquid systems with PMMA was considered, but this was disregarded as the aqueous phase consisted of concentrated salt solutions that were deemed too corrosive. Glycerol solutions were also considered, but the viscosity of these solutions was considered too high when over 70 wt% of glycerol was added, limiting the maximum achievable RI to about 1.430. A number of solids below this limit were then considered including polytetrafuoroethylene (PTFE), polyformaldehyde (PFA), polychlorotrifuoroethylene (PCTFE), and silicone rubber, with respective RIs of 1.380, 1.410, 1.430, and 1.420. Silicone rubber was ultimately selected as this was fairly transparent and chemically inert. The silicone rubber was then RI matched to two immiscible fuids, where the aqueous phase was a 67.9 wt% glycerol solution and the organic phase was methylcyclohexane. In another study, by Stöhr et al.  $(2003)$  $(2003)$  $(2003)$ , two-phase liquid flows through porous media were investigated using PLIF and a fully matched RIM system. The porous medium consisted of fused quartz matched to two immiscible fuid combinations. The frst immiscible fuid was a mixture of silicone oils (98 wt% Dow Corning 556 fuid and 2 wt% Dow Corning 200 fuid), while the second fuid was an aqueous solution of 58 wt% zinc chloride  $(ZnCl<sub>2</sub>)$ .

Given that the majority of liquid–liquid RIM systems reported in Tables [5](#page-20-0) and [6](#page-23-0) have RIs around 1.40, it is surprising that ETFE (1.403) has not been reported in the literature as a matching solid for these systems. A compatible non-hazardous and readily available solid–liquid–liquid RIM system can be created using ETFE with a silicone oil and an approximately 50 wt% glycerol solution. Since silicone oils have a wide range of viscosities with RIs near 1.400, this system allows for considerable tuning of the liquid viscosity ratios. Figure [10a](#page-23-1) shows an ETFE pipe in air, while Fig. [10](#page-23-1)b shows the same ETFE pipe matched to a stratified colourless liquid–liquid system of a  $10^{-5}$  m<sup>2</sup>/s silicone oil (on top) and a 51 wt% glycerol solution (on the bottom). Viewed face-on, the interface between the two liquids is barely visible in Fig. [10b](#page-23-1); however, the top silicone oil layer light undergoes signifcantly more visible colour splitting along the length of the scale.

### <span id="page-22-1"></span>**4 Refractive index, density, and viscosity tuning**

While some liquids are naturally close optical matches to other liquids or solids, the RI of liquids in general can to be tuned to obtain a required RI matching degree by mixing miscible liquids or adding soluble solids. This practice also requires the ability to predict the RI of the resulting liquid mixtures or solutions. The RI of such mixtures can be predicted by numerous relations, with the most common of these listed below; here: *n* is the RI,  $\varphi$  is the volume fraction and the subscripts indicate the component.

A simple prediction of the RI of a multicomponent liquid can be made using the empirical Arago-Biot (AB) equation (Arago and Biot [1806](#page-32-35); Reis et al. [2010](#page-36-35)), which is based on linear volumetric additivity for each component in the mixture:

$$
n = \sum_{i} \varphi_i n_i,\tag{4}
$$

and is similar to the empirical Gladstone–Dale (GD) equation (Gladstone and Dale [1863;](#page-33-41) Sharma et al. [2007](#page-37-36)):

$$
n-1 = \sum_{i} \varphi_i(n-1). \tag{5}
$$

Another empirical relation is the Lichtenecker (L) or Lichtenecker–Rother equation (Lichtenecker [1926](#page-35-28); Lichtenecker and Rother [1931](#page-35-29); Heller [1945\)](#page-34-39) which, as Simpkin [\(2010](#page-37-37)) showed, in fact, has a theoretical basis and can be derived from Maxwell's equations:

$$
\ln n = \sum_{i} \varphi_i \ln n_i, \tag{6}
$$

while relatively common Newton (N) equation (Newton [1704;](#page-36-36) Kurtz and Ward [1936](#page-35-30); Reis et al. [2010\)](#page-36-35) also has a theoretical foundation:

$$
n^2 = \sum_i \varphi_i n_i^2. \tag{7}
$$

$\boldsymbol{n}$	Phase	Material	Density $(kg/m3)$	Dynamic viscosity* (mPa s)	Interfacial tension (mN/m)	References
1.422	Sol.	Silicone rubber			n/a	Burdett et al. (1981)
	Org.	Methylcyclohexane	772		41.3	
	Aq.	67.9 wt% Glycerol 32.1 wt% Water	1173	19.3		
1.460	Sol.	Fused quartz	2200	n/a	n/a	Stöhr et al. (2003)
	Org.	98 wt% Dow Corning 556 fluid (silicone oil) 2 wt% Dow Corning 200 fluid (silicone oil)	970	21		
	Aq.	58 wt% Zinc chloride 42 wt% Water	1700	$\overline{4}$		

<span id="page-23-0"></span>**Table 6** List of selected solid–liquid–liquid RIM systems

\* Temperature for viscosity is provided when stated; otherwise, it is assumed that this is at ambient laboratory conditions between 20 and 25 °C



**Fig. 10** ETFE ( $RI = 1.403$ ) pipe in: **a** air, demonstrating large optical distortions; and **b** stratified layers of refractive-index-matched liquids  $(RI = 1.399)$  comprising a silicone oil on top and a 51 wt% glycerol solution on the bottom

<span id="page-23-1"></span>Proceeding now to more complex models, the more involved theoretical Oster (O) equation (Oster [1948](#page-36-37); Sharma et al. [2007](#page-37-36)) is:

$$
\frac{(n^2-1)(2n^2+1)}{n^2} = \sum_{i} \varphi_i \frac{(n_i^2-1)(2n_i^2+1)}{n_i^2},
$$
 (8)

while the Lorentz–Lorenz (LL) equation, which is also a theoretical model that is based on material polarizability (Lorentz [1906;](#page-35-31) Pacák and Kodejš [1988](#page-36-38)), has a similar form:

$$
\frac{n^2 - 1}{(n^2 + 2)} = \sum_{i} \varphi_i \left( \frac{n_i^2 - 1}{n_i^2 + 2} \right),\tag{9}
$$

as does the empirical Eykman (E) equation (Eykman [1895](#page-33-42); Dreisbach [1948](#page-33-43); Sharma et al. [2007](#page-37-36)):

$$
\frac{n-1}{(n+0.4)} = \sum_{i} \varphi_i \left( \frac{n_i^2 - 1}{n_i + 0.4} \right).
$$
 (10)

Furthermore, the RIs of binary mixtures can be calculated by the Eyring–John (EJ) equation (Eyring and Jhon [1969](#page-33-44)):

$$
n = n_1 \varphi_1^2 + 2(n_1 n_2)^{1/2} \varphi_1 \varphi_2 + n_2 \varphi_2^2, \tag{11}
$$

and for dilute binary mixtures, where component "2" is denoted here as the dilute component, the RI can be calculated using the theoretical Wiener (W) equation (Wiener [1910](#page-37-38); Heller [1945;](#page-34-39) Wiederseiner et al. [2011](#page-37-12)):

$$
\frac{n-1}{n^2+2n_1^2} = \varphi_2 \frac{n_2^2 - n_1^2}{n_2^2 - 2n_1^2},\tag{12}
$$

or using the theoretically derived Heller (H) equation (Heller [1945](#page-34-39); Mehra [2003](#page-35-32)):

$$
\frac{n - n_1}{n_1} = \frac{3}{2}\varphi_2 \frac{n_2^2 - n_1^2}{n_2^2 - 2n_1^2}.
$$
\n(13)

In all the above expressions, the volume fraction of a component " $i$ ",  $\varphi_i$ , is defined using Eq. [14,](#page-23-2) where *V* and *x* are molar volumes and molar fractions, respectively (Sharma et al. [2007](#page-37-36)):

<span id="page-23-2"></span>
$$
\varphi_i = \frac{x_i V_i}{\sum_j x_j V_j}.\tag{14}
$$

The theory behind the RI of liquid mixtures is discussed by Reis et al. ([2010\)](#page-36-35), who explains that differences between the AB and N equations, which are amongst the simplest of the above relations, stem from the rigorous defnitions of RI before and after mixing, respectively. In trying to establish which relation appears better suited for the prediction of the RI of liquid mixtures, we examined comparisons performed by a number of authors. In one such effort that focussed on binary mixtures, Tasić et al. ([1992\)](#page-37-39) compared the AB, GD, LL, W, and H equations concluding that LL predictions agreed very well with RI measurements, whereas the AB relation was found to be the poorest predictor. Mehra [\(2003](#page-35-32)) tested the GD, LL, W, and H equations in relation to binary mixtures of hexadecane and heptadecane with a series of alcohols, and concluded that the GD and W equations did not perform as well as the LL or H equations. The authors also noted that the GD equation gave the same results as the W equation in very dilute mixtures but that neither worked well at higher concentrations. In their extensive study, Sharma et al. ([2007\)](#page-37-36) compared the AB, GD, N, O, LL, E, EJ, W, and H equations, and considered the H equation to be most accurate for their specifc binary test mixtures of eucalyptol with hydrocarbons. The authors noted small deviations between the AB and GD predictions, and reported that the W equation exhibited larger deviations from experimental RI values than the E, EJ, and H equations. In another comprehensive study involving multiple empirical and theoretical relations, Mandava et al. [\(2015](#page-35-33)) compared the AB, GD, N, O, LL, E, EJ, W, and H equations and found that the O relation was the worst performing for their mixtures, while the N equation performed the best. Isehunwa et al.  $(2015)$  $(2015)$  compared predictions of the RIs of selected binary mixtures from the AB, GD, LL, W and H equations, and found that their own modifed AB equation had the widest temperature applicability.

From the above variability over the best performing relation(s), but also a large number of similar studies in the literature, it is clear that these equations should be treated as a guide and that RI should be measured whenever possible, but also that it is a great challenge to identify one relation that can be generally accepted as being best with different relations performing better with different liquid phases and their relative mixture concentrations. Nevertheless, Mehra ([2003\)](#page-35-32) states that the most frequently used equation is LL equation, and although in the above comparisons only, Tasić et al. ([1992\)](#page-37-39) and Mehra [\(2003](#page-35-32)) found LL to be the most accurate approach, none found that it was the worst performing. Based on this observation, the present authors consider that the LL equation appears to be the most promising, at least as a good starting point, for the purposes of RI tuning.

Importantly, combining three or more liquids or additives allows for the matching or tuning of parameters over and above the RI, i.e., density, viscosity, surface, and interfacial tensions, for the purpose of fow similarity. Density tuning is desirable when gravitational effects need to be considered in both solid–liquid and liquid–liquid systems and is particularly important when trying to establish neutrally buoyant solids (Bailey and Yoda [2003\)](#page-32-22). Viscosity tuning is important when trying to obtain direct matches to actual liquids or when matching Reynolds numbers. Likewise, the interfacial tension can be important in liquid–liquid systems when investigating interfacial phenomena (droplet breakup, atomization, etc.). The prediction of density, viscosity, and surface tension is beyond the scope of this paper, so the reader is referred to Polling et al. ([2001](#page-36-39)) which is an excellent reference source covering a range of methods for estimating these properties for a wide range of liquids, and their mixtures. Combinations can also allow RI tuning at two wavelengths simultaneously, for example, the wavelengths of laser emission and dye fuorescence (Saksena et al. [2015\)](#page-37-33).

Furthermore, many physio/biological, petrochemical, and other flows involve complex non-Newtonian fluids for which it is often desirable to tune additional properties in RIM experiments, such as the fuid rheology, to achieve dynamic similarity with fows of interest. For example, the shear thinning and viscoelastic behaviour of blood is often modelled by adding xanthan gum, and the further addition of NaI or sodium thiocyanate (NaSCN) can be used to reduce the viscosity of the resultant xanthan gum mixtures (Najjari et al. [2016\)](#page-36-40).

Tuning for dynamic similarity in two-phase fow systems where (Newtonian) viscosity, density, and interfacial tension are important can be achieved by satisfying Eqs. [15](#page-24-0), [16](#page-24-1), [17](#page-24-2) for the liquid phases of interest A and B, and the surrogate/model liquids SA and SB (Saksena et al. [2015](#page-37-33)). Equations [15](#page-24-0) and [16](#page-24-1) equate the density and viscosity ratios of the surrogate and actual liquids, while Eq. [17](#page-24-2) is required for matching dimensionless numbers concerning interfacial tension including the Weber number, Bond number, capillary number, Eötvös number, and Ohnesorge number:

<span id="page-24-0"></span>
$$
\frac{\rho_{SA}}{\rho_{SB}} = \frac{\rho_A}{\rho_B};\tag{15}
$$

<span id="page-24-1"></span>
$$
\frac{\mu_{SA}}{\mu_{SB}} = \frac{\mu_A}{\mu_B};\tag{16}
$$

<span id="page-24-2"></span>
$$
\frac{\sigma_{\text{S}}^3 \cdot \text{S}\text{B}}{\mu_{\text{S}}^4} = \frac{\sigma_{\text{A}:\text{B}}^3 \rho \text{A}}{\mu_{\text{A}}^4}.
$$
\n(17)

A highly tuneable liquid–liquid RIM system with a RI covering the range from 1.395 and 1.436 was created by Saksena et al. ([2015\)](#page-37-33). This system simultaneously allowed control over RI, viscosity, and density, as well as a potential 4th parameter. This fexibility was achieved through having two-liquid-mixture phases: an aqueous solution containing 1,2-propanedoil and caesium bromide (CsBr), and an organic phase comprising a light (5  $\times$  10<sup>-6</sup> m<sup>2</sup>/s) and heavy (5  $\times$  10<sup>-5</sup> m<sup>2</sup>/s) silicone oil blended with 1-bromooctane. The system was extended for wider viscosity and density ratios by Cadillon et al. [\(2016](#page-32-31)) by replacing the silicone oils with ones with even larger viscosity differences  $(1 \times 10^{-6} \text{ m}^2/\text{s} \text{ and } 5 \times 10^{-3} \text{ m}^2/\text{s})$  to obtain a system with an RI in the range 1.382–1.436.

An experimental system will typically defne a working temperature range. Temperature variations can, however, give rise to RI variations, meaning that RI tuning may be required even for what superficially appears to be a RIM system. In one example, Fort et al. ([2015\)](#page-33-17) demonstrated that para-cymene closely matched PMMA at 291.7 K, but noted that a cooling system may be required if heat sources, such as pumps, exist within a flow loop. It was, therefore, suggested that para-cymene's RI could be tuned through adding a small amount of cinnamaldehyde with an RI of 1.562 to allow RI matching with PMMA at higher temperatures.

The present authors measured the RI of two liquids over a range of temperatures: (i) polydimethylsiloxane silicone oil with a kinematic viscosity  $10^{-5}$  m<sup>2</sup>/s; and (ii) four RIM glycerol solutions, and the results are plotted in Fig. [11.](#page-25-0) The two fuid types have different RI temperature gradients, which can cause matched systems to diverge with temperature. A change of about 4 °C corresponds to an RI change of the same magnitude as that due to a 1 wt% difference in glycerol concentration. These variations can, in principle, be limited using ternary systems to simultaneously tune the RI and the RI temperature gradients of a two-liquid system; however, no system has been found in the literature implementing this RI temperature gradient matching technique.

The effect on the RI of different glycerol solution concentrations over the entire range of mole fractions for temperatures between 25 and 60 °C is reported in a comprehensive study by Leron et al. [\(2012](#page-35-34)), and associated results are plotted here in Fig. [12](#page-25-1). This fgure shows that the temperature change of 35 °C corresponds to a change in RI of up to  $\sim 0.01$ .

The following sections describe organic and aqueous liquids, as well as salts found in experiments featuring RIM systems.

### **4.1 Organic liquids**

A wide range of organic liquids have been employed in RIM systems, including pure hydrocarbons, hydrocarbon mixtures, mineral or silicone oils, fuorocarbons, etc. A selection of such fuids is listed in Table [7](#page-26-0), covering an RI range from 1.251 to 1.631. The table forms a guide for the



<span id="page-25-0"></span>**Fig. 11** RI variation with temperature for a silicone oil with viscosity 10−<sup>5</sup> m2 /s and a range of closely RIM glycerol solutions



<span id="page-25-1"></span>**Fig. 12** RI variation with temperature between 25 and 60 °C for a full range of glycerol solutions; data from Leron et al. [\(2012](#page-35-34))

selection of potential components for tuning organic mixtures. For instance, high RI organic chemicals like tetralin  $(RI = 1.541)$  can be added to increase the RI of another organic phase, while similarly, low RI liquids like tetradecafuorohexane can be added to lower the RI. Table [7](#page-26-0) also provides water solubility as well as PMMA compatibility information that has been obtained principally via supplier datasheets and should be regarded as approximate guidance only.

In general, the RI of hydrocarbons within a given chemical class generally increases with the size/weight or complexity of their chemical structure. Fluorocarbons have relatively low RIs, and importantly, many fuorocarbons have RIs that are lower than water. This proximity in their RI to that of water makes fuorocarbon-based blends suitable as



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**Table 7** continued

\* Temperature for viscosity is provided when stated; otherwise, it is assumed that this is at ambient laboratory conditions between 20 and 25 °C

organic liquid candidates for solid–liquid–liquid RIM systems with water and FEP as the aqueous and solid phases, respectively. Although sourcing fuorocarbons in both reasonable quantities and cost may, in some cases, prove dif-ficult, especially for large system (Saksena et al. [2015](#page-37-33)), Hibberd et al. [\(2007](#page-34-36)) reported using tetradecafuorohexane and n-hexane with RIs of 1.251 and 1.378, respectively, to prepare a water-based RIM emulsion. Of importance in this case is the fammability and relatively low boiling points of tetradecafuorohexane (<61 °C) and *n*-hexane (<69 °C), which introduce the need to apply caution when handing these fuids outside of controlled, closed environments. Longer chain fuorocarbons are potentially even more suitable for experimental systems, e.g., Vitreon (perfuoroperhydrophenanthrene) has  $RI = 1.335$  (close to water) and is both non-toxic and non-volatile (Georgalas et al. [2011](#page-33-45)). A combination of FEP, water and perfuoroperhydrophenanthrene, therefore, appears to be a promising candidate for a water-based RI matched liquid-liquid system.

Silicone oils are another group of organic liquids worth mentioning. In general, they are non-toxic and have wide ranges of both RIs and viscosities which make them ideal candidates for tuning the viscosity of organic mixtures. Dow Corning 200 silicone (polydimethylsiloxane) oils, which are the most common silicon oil family, cover kinematic viscosities in the range  $6.5 \times 10^{-7}$ –1 m<sup>2</sup>/s, but have associated RIs in the relatively narrow range 1.375–1.404, as shown in Fig. [13.](#page-28-0) Viscosity and RI tuning can then be achieved through mixing with a range of available miscible higher RI silicone oils like Dow Corning 550 ( $RI = 1.490-$ 1.500, viscosity  $1.0 \times 10^{-4}$ –1.50 ×  $10^{-4}$  m<sup>2</sup>/s), Dow Corning 556 (RI = 1.46, viscosity 2.25 × 10<sup>-5</sup> m<sup>2</sup>/s), or Dow Corning 710 (RI = 1.533, viscosity 5  $\times$  10<sup>-3</sup> m<sup>2</sup>/s).



<span id="page-28-0"></span>**Fig. 13** Variation of RI with viscosity for Dow Corning 200 (polydimethylsiloxane) silicone oils (Dow Corning datasheets)

Finally, solubility in water (indicated in Table [7](#page-26-0)) and in other organic solvents should be checked when considering a liquid–liquid RIM system. Hassan and Dominguez-Ontiveros ([2008\)](#page-34-15) matched the RI of soda-lime glass to two different organic systems: (i) diethylphthalate; and (ii) a mixture of isopropanol and methylnaphthalene. Isopropanol, however, is fully soluble in water and, therefore, organic mixtures that include isopropanol cannot be used when water is the aqueous phase in a liquid–liquid system. UCON polyalkylene glycol based oils are also widely found in RIM-based experimental literature, but are again miscible with water.

### **4.2 Aqueous and hydrophilic liquids**

Aqueous systems are comprised of water, which has a relatively low RI (of 1.333). This means that RI matching to the majority of available and commonly used solids and organic liquids, which have considerably higher RIs (hydrogels, FEP and fuorocarbons being exceptions), is a challenge and requires signifcant amounts of tuning with mixing or additives. An increase in the RI of an aqueous phase is often achieved through the addition of glycerol (Bailey and Yoda [2003](#page-32-22); Takamura et al. [2012](#page-37-41)). Glycerol has a relatively high RI (of 1.473), thereby allowing a range of RI values to be spanned based on the water–glycerol ratio. At the same time, the dynamic viscosity of pure glycerol is 1.412 Pa s, so the addition of glycerol to water (0.89 mPa s) is also accompanied by a signifcant increase in viscosity. This increase in viscosity can be undesirable when studying liquid–liquid flows that mimic real flow systems, as the water-to-oil viscosity ratios can be signifcantly altered (Morgan et al. [2013,](#page-36-6) [2016\)](#page-36-7). In such cases, glycerol solutions can be further combined with alcohols in ternary systems (Moreira et al. [2009\)](#page-36-43) or salts, so that density, viscosity, or surface tension can be independently controlled. Baldwin et al. ([1989](#page-32-20)) created a mixture of 79 vol% saturated NaI solution, 20 vol% glycerol, and 1 vol% water which matched the RI of PMMA, while also having approximately the same viscosity of blood.

The density and surface tension of glycerol solutions can also be modifed via the addition of certain alcohols, e.g., ethanol, which has a signifcantly lower surface tension and density ( $\sigma = 23.4 \times 10^{-3}$  N/m;  $\rho = 789$  kg/m<sup>3</sup>), but similar viscosity and RI ( $\mu = 1.2 \times 10^{-3}$  Pa s;  $n = 1.361$ ) compared to water (Vazquez et al. [1995;](#page-37-42) Khattab et al. [2012;](#page-34-43) Mathie et al. [2013;](#page-35-37) Markides et al. [2016](#page-35-38)). Nevertheless, ethanol is known to attack some plastics (e.g., PMMA), so chemical compatibly should be checked at the experiment design stage. Table [8](#page-29-0) lists common liquids suitable for the RI tuning of aqueous solutions, where we have again included PMMA compatibility data that have been collated from supplier datasheets.

<span id="page-29-0"></span>



\* Temperature for viscosity is provided when stated; otherwise, it is assumed that this is at ambient laboratory conditions between 20 and 25 °C

Salts are commonly added to increase the RI of aqueous RIM systems (see Table [9\)](#page-29-1). The ability of salts to increase RI is usually limited by their solubility in the solvent, so the RI of a given solution can be modifed from that of the pure liquid (e.g., water with RI of 1.333) up to that achieved by the saturated salt solution. Nevertheless, this solubility can be altered by further additives, such as crown ethers (Lopez-Gejo et al. [2007](#page-35-39)). On the other hand, the introduction of salts also leads to an increase in the density and viscosity of the resulting aqueous solution, as well as an increase in the risk of corrosion (e.g., electrochemical oxidation of metals), and therefore, care must be taken when selecting the materials to be used in any experimental campaign (e.g., plastics and, in particular, metals). The typical criteria for the selection of a given salt additive are: (i) increase in the RI with concentration; (ii) salt solubility; (iii) increase in the viscosity with concentration; (iv) increase in the density with concentration; (v) compatibility and corrosion potential; and (vi) stability of a given salt solution.

<span id="page-29-1"></span>**Table 9** List of solids used for RI tuning of aqueous solutions

Solubility $(g/100 \text{ g at } 20 \text{ °C})$	$n_{\rm L}$	<b>PMMA</b> compatibility	References
>10	1.330-1.345		Alahyari and Longmire (1994)
35.9	1.330-1.380	Good	Hayes $(2015)$
123 (at $25^{\circ}$ C)	1.330-1.405		Li et al. $(2013)$
74.5	1.330-1.442	Good	Hayes $(2015)$
395	1.330-1.470	Good	Stöhr et al. (2003)
139	1.330-1.480		Budwig $(1994)$
224	1.330-1.490		Jan et al. (1989)
125	1.330-1.490	Good	Prasad et al. $(1991)$
~2000	1.330-1.490	Good	Dijksman et al. (2012)
178	1.330-1.499	Good	Bai and Katz $(2014)$
170	1.330-1.503	Good	Borrero-Echeverry and Morrison (2016)
>1000	1.330-1.550	Good	Dijksman et al. (2012)
432	1.330-1.620	Good	Hendriks and Aviram (1982)

All these criteria are temperature sensitive making temperature control important especially in saturated systems where drops in temperature can lead to salts leaving the solution.

Referring to Table [9](#page-29-1), the present review of the literature has revealed that NaI is the most widely employed salt in RIM experiments. Typical NaI solutions matching the RI of PMMA have concentrations in the range of 60.0–64.8 wt%, yielding a RI in the range 1.485–1.491 (Imao et al. [1996](#page-34-44); Parker and Merati [1996;](#page-36-17) Uzol et al. [2002;](#page-37-16) Mehta et al. [2007](#page-35-16); Uzol et al. [2007;](#page-37-17) Soranna et al. [2008](#page-37-13); Yuki et al. [2008](#page-38-8); Wu et al. [2009](#page-37-20); Amatya and Longmire [2010](#page-32-19); Wu et al. [2011](#page-37-1), [2012](#page-37-21); Yuki et al. [2011;](#page-38-9) Yuan et al. [2012;](#page-38-13) Tomac and Gregory [2014](#page-37-19)). A detailed study of the use of NaI for RI matching (aimed at PIV measurements) over an even wider RI range (1.330–1.510) was performed by Bai and Katz [\(2014](#page-32-28)), who also matched NaI solutions to three optical solids formed from resins with RIs in the range 1.495– 1.508. NaI solutions with RIs >1.499 could only be reached at temperatures >23 °C. Of interest in the context of RI matching is a useful study by Narrow et al. [\(2000](#page-36-24)), who developed a model for predicting the RI of NaI solutions for a known temperature and concentration. At the same time, the addition of NaI to water does not signifcantly affect the liquid solution's viscosity. NaI solutions have kinematic viscosities of ~1.1  $\times$  10<sup>-6</sup> m<sup>2</sup>/s, which is only slightly higher than that of water that has a viscosity of 1.0 <sup>×</sup> <sup>10</sup>−<sup>6</sup> m2 /s at 20 °C (Uzol et al. ([2002,](#page-37-16) [2007\)](#page-37-17). NaI like many iodides undergoes photodissociation, oxidizing in the presence of light (Chen and Fan [1992](#page-33-29)). This can, however, be counteracted through the addition of 0.1 g of sodium thiosulfate (Na<sub>2</sub>S<sub>2</sub>O<sub>3</sub>) per L (Ghatage et al. [2014](#page-33-35); Narrow et al. [2000;](#page-36-24) Parker and Merati [1996\)](#page-36-17). The precipitation of iodide can also be reduced by the addition of 20 mg of ascorbic acid per  $\text{cm}^3$  of salt solution (Jacobs et al. [1988\)](#page-34-33).

NH4SCN is the second most common salt reported in RIM systems and was examined in detail in the context of RIM experiments by Borrero-Echeverry and Morrison [\(2016](#page-32-42)). This study reported an RI value of 1.503 for a 62.6 wt% aqueous-NH<sub>4</sub>SCN solution, also with a relatively low kinematic viscosity of  $1.7 \text{ mm}^2$  s and a density of 1140 kg m<sup>-3</sup>. NH<sub>4</sub>SCN is, like other thiocyanates, toxic if inhaled or ingested and it is important that it is handled with care and suitable protective clothing.

Hendriks and Aviram [\(1982](#page-34-10)) investigated the use of  $ZnI_2$  in RIM systems. It was shown that  $ZnI_2$  can form aqueous solutions with a high RI (1.620) at 81 wt%, with a kinematic viscosity just over  $5 \times 10^{-6}$  m<sup>2</sup>/s at 21 °C, and that for a less concentrated 60 wt% solution, the viscosity falls to  $2 \times 10^{-6}$  m<sup>2</sup>/s whilst matching the RI of PMMA at 1.490. Hence,  $ZnI_2$  solutions have the potential to be utilized as aqueous RIM systems matching all solids from FEP to PS in RI. Nevertheless,  $\text{ZnI}_2$  like many salts is

hydroscopic and the accurate measurement of the weight of a given salt requires the salt to be dried frst (Hendriks and Aviram [1982](#page-34-10)).

The material compatibility and corrosion issues associated with the use of salts have been addressed by a number of investigators. Cenedese and Viotti ([1996\)](#page-33-28) noted that  $ZnCl<sub>2</sub>$  corrodes aluminium and so opted instead to use glycerol in their RIM experiments. Similarly, Reddy et al. [\(2013](#page-36-21)) considered a 55 wt% NaI solution too corrosive, while a solution of 42 wt% KSCN and a solution of 45% NH4SCN were both considered too corrosive and too toxic. Borrero-Echeverry and Morrison ([2016\)](#page-32-42) found that NH4SCN solutions were compatible with 6061 aluminium alloy, anodized aluminium, 316 stainless steel, common plastics, and glass but corroded plain steel and 304 stainless steel. Furthermore, Stöhr et al. ([2003\)](#page-37-15) showed that a 58 wt% concentration of  $ZnCl<sub>2</sub>$  has a pH of about 2 and is, therefore, incompatible with PMMA and many fuorescent dyes. Bailey and Yoda ([2003\)](#page-32-22) were successful in creating a ternary mixture of NH4SCN, water, and glycerol that was compatible with plastics including PMMA, PC, and PVC. However, it corroded many metals, with the exception of 304 stainless steel and some aluminium alloys. In a couple of other approaches of interest; Dijksman et al. ([2012\)](#page-33-10) noted that sodium polytungstate is a relatively non-toxic salt, forming solutions with RIs up to 1.550 which are compatible with most plastics and metals, with the exception of aluminium, while Prasad et al. ([1991\)](#page-36-14) matched sodium salicylate solution to PMMA in static conditions. Sodium salicylate, however, was not deemed suitable for flow experiments, as it required high concentrations, was expensive and changed colour over time.

### <span id="page-30-0"></span>**5 Conclusions**

This review paper presented a number of previously employed refractive-index-matched experimental systems (solid and liquid combinations) reported in the literature, covering more than 280 references. Although the primary focus is on optical properties, information is included on broader experimental design aspects, including safety, toxicity, material compatibility, the role of temperature, wavelength, solubility/miscibility, as well as the use of liquid mixtures and/or additives (e.g., salts) for the tuning/adjustment of properties such as density, viscosity, and surface/ interfacial tension. The practice of refractive-index matching forms an important component of the accurate and reliable application of optical experimental methods such as direct photography, laser-induced fuorescence, laser Doppler velocimetry or phase Doppler anemometry, particle image or tracking velocimetry (and variants thereof), and others, to a variety of multiphase fows that include

solid–liquid, liquid–liquid, and solid–liquid–liquid systems, with similar issues applying to internal or external single-phase (liquid) flows in confined solid spaces or over solid obstacles. This overview has shown that aqueous and organic phases can, in principle, be matched over a wide range of refractive indices (1.330–1.620), as well as varying viscosities, densities, and even surface and interfacial tensions. Further refractive-index tuning, through mixing of liquids or the addition of salts into solutions, can also be used for optical matching with improved accuracy, and can also allow the simultaneous tuning of other desirable properties, such as viscosity and density, for the purpose of attaining fow similarity. In the context of employing optical measurement methods, including advanced opticalbased flow diagnostic techniques, the use of these refractive-index matching techniques is important in minimizing distortions in either illumination and/or detection caused by refractive-index differences between individual phases, increasing the overall accuracy of the measurements, as well as allowing for measurements in more complex (e.g., dispersed liquid–liquid) interfacial fows.

The liquid–liquid system matches reported in the literature were found to be concentrated near a refractive index of  $\sim$  1.40, predominantly using silicone oils as the organic phase and glycerol–water solutions as the aqueous phase. With regard to solid materials, borosilicate glass and PMMA were found to be the most commonly employed solids in refractive-index-matched experiments, but these have high refractive indices (1.474 and 1.490, respectively) compared to water (1.333). Lower refractiveindex-materials have been employed, such as FEP (1.338) and silicone rubbers (1.410–1.440), but these are less common. In particular, FEP and hydrogels are close matches for pure water, while silicone rubbers are a good option when creating fexible (compliant) models of biological or other structures. The use of solids with refractive indices between 1.333 and 1.410 is rare, but desirable, as this does not necessitate the use of additives to the aqueous phase for refractive-index tuning and matching. Suitable solids that can optically match the aforementioned liquid–liquid systems do exist, including for example fuoropolymers such as ETFE (1.403); however, we have not identifed instances of the use of such plastics being reported within the literature. Therefore, an especially promising, compatible, nonhazardous, and readily available solid–liquid–liquid RIM system matched at ~1.40 uses ETFE, a silicone oil, and a  $~50$  wt% glycerol solution. Importantly, this system allows tuning of the liquid viscosity ratios due to the wide range of viscosities of silicone oils at RIs of ~1.40, and the ability to vary the glycerol–water mixing ratio. A second solid–liquid–liquid combination is that of FEP, water, and perfuoroperhydrophenanthrene, which is based on pure substances rather than mixtures. This relatively unusual combination is of particular interest, since the RIs of its three phases are all at or close to that of water (1.33–1.34), that in fact it includes. Depending on the accuracy required, this combination may not require any further tuning (mixing, additives) or adjustment (e.g., by temperature or otherwise). To the best of the authors' knowledge, neither of these two solid/liquid combinations have been previously reported in experimental multiphase flow investigations.

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### **Compliance with ethical standards**

**Confict of interest** SFW is an employee of Cameron, a Schlumberger company. IZ is an employee of Dantec Dynamics.

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