Chapter 6
Experimental Determination of Rheological Behavior

This chapter will be divided into two main parts. The behavior of alloys during partial solidification will be considered first but only alloys with a globular morphology will be examined. In this case, the alloy is considered as a homogeneous medium and therefore, the viscosity only (and sometimes the fluidity) has been determined. Alloys during partial remelting will thereafter be considered. In this case, the solid fraction can vary in large proportions so that the two approaches mentioned above will be detailed. In each part, the experimental methods will be examined first and then, the experimental results will be presented. It is necessary to mention here that only the most representative results will be presented and not all of the results in the literature.

In addition to these two main parts, a comparison between partially solidified and partially remelted alloys will be performed. This comparison is important in view of the latest industrial developments of the rheocasting processes. Finally, the question of yield stress will be addressed.

6.1 Partially Solidified Alloys

6.1.1 Experimental Methods

Rotation and Translation Viscometer

The Couette viscometer test has been used since the beginning of research concerning semisolid alloys for their rheological characterization [1, 2]. The advantage of this test is the possibility to generate the globular microstructure suitable for the forming operation directly in the apparatus. During the test, the alloy is sheared between two concentric cylinders, the inner cylinder being fixed and the outer cylinder rotating at constant angular velocity. A fixed outer cylinder and a rotating inner cylinder define a Searle viscometer. The torque, which is required to shear the alloy is then continuously recorded. From the angular velocity and the torque, it is possible to deduce the shear rate and the shear stress after some simple

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assumptions. The test is therefore very simple, but problems occur as soon as very clean experiments are required to produce quantitative data. The problems are concerned with:

- The nature of the materials used to build the apparatus owing to the aggressive behavior of particular alloys. This problem is particularly important in the case of aluminum alloys which dissolve steels. This requires either protective coatings on steels or the use of other materials like graphite.
- The control and homogeneity of the temperature: This aspect is particularly important in the case of semisolid alloys for which the viscosity is strongly dependent on solid fraction and therefore on temperature. Problems associated with temperature can have three origins: temperature control, temperature gradients in the device, and adiabatic heating of the alloy during shearing. The control of the temperature is usually carried out through one or several thermocouples, which are positioned as close as possible to the alloy, which can lead to a small difference with the temperature of the alloy. In a similar way, longitudinal temperature gradients can be present, which directly depend on the heating system. Viscous heating can be generated in the case of high shear rates. However, for aluminum alloys sheared at angular velocity of 500 rpm, it has been demonstrated that this heating does not exceed a few thousandths of a degree which is completely negligible [3].
- Turbulence effects: The equations which allow the determination of the viscosity in the viscometer assume laminar flow for the semisolid alloy. This assumption, which may be wrong in the case of liquid sheared at high velocities, is generally valid in the case of semisolid alloys for which the viscosity increases rapidly with increasing solid fraction.
- End effects: The viscometer has a finite length so that the torque must in principle be corrected to account for end effects. Indeed, a small part of the alloy is present under the inner cylinder and it is sheared under a variable velocity lower than that applied in between the cylinders. For alloys, which exhibit a strong shear thinning behavior, this shearing can induce a relatively important torque, all the more when the length of the viscometer is small.
- Wall slip and groove effects: The shear that is imposed on the alloy located between the two cylinders is assumed to vary linearly from the wall of the fixed cylinder to that of the rotating one. The alloy in contact with the rotating wall is therefore moving with the same velocity. However, slip can occur at this wall, which can lead to erroneous measurements. To avoid slip, longitudinal grooves are often machined along the two cylinders, which can cause nondesirable effects.
- Effects of variation of solid fraction: Good measurements assume that the material, which is sheared between the cylinders is homogeneous. This assumption is certainly valid in the case of a liquid, but it does not hold when particles are present as is the case for an alloy during solidification. It is, however, quite reasonable for particles that are small compared with the dimension of the gap between the cylinders. It must be noted that gravity or centrifugal acceleration can lead to heterogeneous distribution of the particles.

– Inertia effects: These effects can happen during rapid changes of shear rate and lead to some delay in the torque measurement. Extrapolation of curves is therefore necessary to obtain the material response at constant structure and this can lead to some error.

Beside the Couette viscometer, other rotation viscometers can be used: the cone–plane viscometer, the plane–plane viscometer or more simply viscometers which are constituted of a part rotating in the solidifying alloy. In this latter case, calibration with fluid of known viscosity is mandatory owing to the complicated geometry of the system.

In addition to rotation shear experiments, shear can be produced by translation of one part of the specimen with respect to the other. This test has been used for the first experiments dealing with the rheological behavior of semisolid alloys in order to establish relations between the strength of the alloy and its tendency to hot crack formation [4,5]. It consists of translating one part of the sample, the other part being fixed. Shear then occurs in a very narrow region so that it is very difficult to know precisely the imposed strain. This test has been taken up again more recently on Al–Si alloys [6] and even more recently on Al–Si–Cu alloys [7]. Another possible configuration of the translation shear test is the use of two concentric cylinders with circumferential grooves. As in the Couette viscometer, the alloy is located in between the cylinders but a translation is imposed to one cylinder with respect to the other. For small displacement, pure shear is imposed on the alloy over a known distance so that strain is also known, provided that no slip occurs at the wall. This apparatus has been used for the study of aluminum alloys during solidification at particularly large solid volume fractions [8]. In the following, the results obtained with these viscometers will not be reported since they are mainly concerned with dendritic structures during solidification and transitions in the behavior when the solid fraction increases.

Shearing of a semisolid alloy can be produced by parallel plate compression. Although this type of test is better suited for partially remelted billets, it has been employed also with billets transferred from the cooling system to the parallel plate test machine [9, 10]. In these experiments, a dead weight is applied on the specimen and the displacement is recorded continuously, the billet temperature being kept constant by using a furnace installed on the press.

Fluidity Tests

Fluidity tests have been used to characterize the rheological behavior of semisolid alloys particularly at relatively low solid fractions. Fluidity does not have a mathematical definition, in contrast with viscosity. It is used by foundry-men and consists of the measure of the length that an alloy travels in a tube of a given geometry before it solidifies. This is therefore a comparative method, which can be carried out for the liquid as well as for the semisolid alloy. Two systems are usually used but it is possible to imagine other ones. The first (Fig. 6.1a) is constituted of a horizontal spiral and a vertical sprue. The liquid or semisolid alloy is poured inside the sprue,

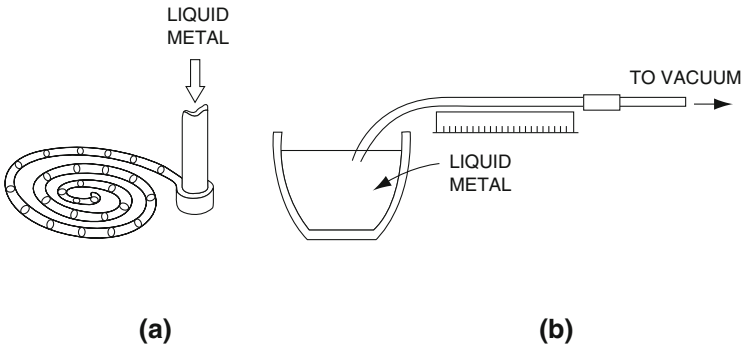

Fig. 6.1 Devices used to measure the fluidity. (**a**) spiral; (**b**) vacuum suction

which travels a given distance in the spiral. In the second apparatus (Fig. 6.1b), the alloy is held in a crucible and a tube connected with a vacuum system is submerged in the alloy. In this case too, the distance that the alloy has traveled in the tube is recorded as the fluidity parameter.

6.1.2 Results Concerning Viscosity

The first results concerning viscosity of alloys during solidification have been obtained at MIT with a Sn–Pb alloy [1]. These authors measured the apparent viscosity of the alloy during solidification with mechanical stirring. The results showed that the stirred semisolid alloy at solid fractions greater than 0.2 behaves like a non-Newtonian fluid with a much smaller viscosity than the same unstirred alloy. This first observation is at the origin of the very large number of studies dealing with the rheology of stirred alloys. The very important work of Joly and Mehrabian [2] on the Sn–Pb system too is among these studies. Today, although numerous studies have been carried out on other systems and have tried to understand more complex aspects of the rheological behavior, these works are still the basis of our knowledge of this behavior. Three main aspects of the variation of the apparent viscosity of these alloys have been studied:

– The variation of the viscosity at constant shear rate during continuous cooling
– The variation of viscosity at constant temperature as a function of the shear rate
– The thixotropic behavior of the alloy, i.e., the time dependence of the rheology

The determination of the apparent viscosity of an alloy during continuous cooling can be viewed as a very simple and fast characterization method of the rheology. The test is carried out at constant shear rate so that the increase of viscosity is due to the increase of the solid fraction. The effects of the shear rate and of the cooling rate can also be studied during these experiments. The classical curves giving the variation of the viscosity as a function of the solid fraction show that the increase

of the shear rate increases the critical solid fraction at which the viscosity increases sharply (Fig. 6.2). Similar curves have been obtained on other semisolid alloys like industrial Al–Si alloys and on alloys containing SiC particles.

In order to express the influence of the solid fraction f_s on the viscosity η of the alloy, laws of the type $\eta = A \exp(Bf_s)$ have been used. Table 6.1 gives the values of B deduced from viscometer experiments on various alloys. In this table,

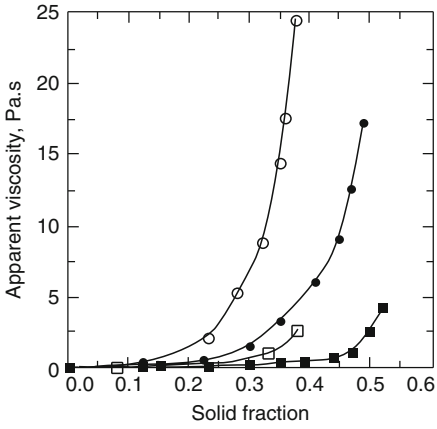

Fig. 6.2 Apparent viscosity as a function of the solid fraction for an A356 alloy sheared at (*filled square*) 27 s^{-1}, (*open square*) 54 s^{-1}, (*filled circle*) 108 s^{-1}, and (*open circle*) 216 s^{-1} during continuous cooling in the solidification interval at 1.2°C min^{-1} (from [11])

Table 6.1 Values of B in the expression: $\eta = A \exp(Bf_s)$ for various alloys

Alloy	Conditions of the tests	Value of B	References
Sn-15%Pb	Continuous cooling at 0.33°C min^{-1}	7.4	[2]
	Shear rate of 750 s^{-1}		
Sn-15%Pb	Continuous cooling at 25°C min^{-1}	17.6	[2]
	Shear rate of 750 s^{-1}		
A356	Continuous cooling at 1.2°C min^{-1}	15	[11]
	Shear rate of 27 to 216 s^{-1}		
A356	Isothermal holding	11.3	[11]
	Shear rate of 27 to 216 s^{-1}		
A356	Continuous cooling at 15°C min^{-1}	10.7	[3]
	Shear rate of 0.063 s^{-1}		
A356	Continuous cooling at 15°C min^{-1}	15.6	[3]
	Shear rate of 105 s^{-1}		
A356 + 18% SiCp	Continuous cooling at 15°C min^{-1}	16.9	[3]
	Shear rate of 105 s^{-1}		
A356	Isothermal holding	9.5	[3]
	Shear rate of 1.6 to 105 s^{-1}		
A356 + 18% SiCp	Isothermal holding	11.7	[3]
	Shear rate of 26 to 210 s^{-1}		

values deduced from isothermal experiments are also included together with results on composites.

The results show a very strong dependence of the viscosity on the solid fraction with B values ranging between 10 and 18. Although it is difficult to derive clear tendencies, it seems that the B value is smaller for low cooling rates and for isothermal holding, which can be considered in fact as a cooling at zero rate. The interpretation that can be put forward for these large B values is the presence of agglomerates of solid globules, which entrap part of the liquid. The liquid fraction participating in the flow can be therefore considerably reduced and this occurs all the more when the agglomerates are formed at high cooling rates.

A similar type of law was derived [10] for the influence of the aspect ratio AR of the globules on the viscosity of A356 Al–Si alloys deduced from parallel plate compression viscometry at shear rates lower than $0.01\,s^{-1}$. For a solid fraction of 0.36, they found that the viscosity increases exponentially with increasing aspect ratio according to: $\eta = K \exp(11.28\,AR)$ for values of AR ranging from 1.4 to 1.8. This law corresponds to a B coefficient ranging from about 44 for AR = 1.4 to about 56 for AR = 1.8. These values are much greater than the values given in Table 6.1, which can be possibly explained by the very low strain rates applied during these compression experiments for which agglomeration of the solid globules is important.

Shear experiments during isothermal holding allow a more precise rheological characterization. They consist of a partial solidification of the alloy at a given shear rate, until a steady shear stress is reached. Then, another shear rate is applied and the response in terms of shear stress is recorded. The main characteristic feature of these experiments is that the measure is performed after sufficient time to reach steady state. This time can be quite long as shown in Fig. 6.3 obtained on an Al–Si alloy A356 [11].

Similar experiments have been carried out by Kattamis and Piccone [12], on an Al–4.5%Cu–1.5%Mg, but at various solid fractions. Figure 6.4 shows the variation

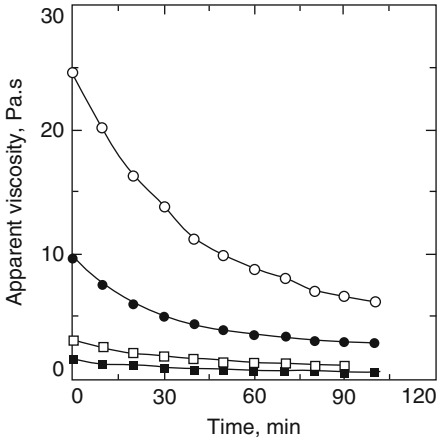

Fig. 6.3 Apparent viscosity as a function of shearing time at 590°C of a A356 alloy ($f_s = 0.35$) partially solidified with a shear rate of (*filled square*) $27\,s^{-1}$, (*open square*) $54\,s^{-1}$, (*filled circle*) $108\,s^{-1}$, and (*open circle*) $216\,s^{-1}$ (from [11])

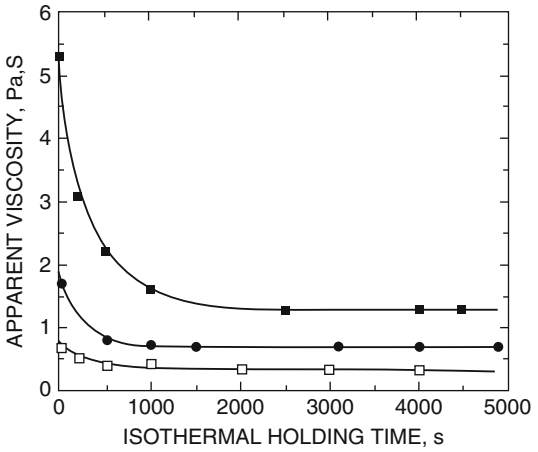

Fig. 6.4 Apparent viscosity as a function of shearing time at a shear rate of $200\,s^{-1}$ and at various temperatures corresponding to solid fraction $f_s = 0.3$ (*open square*), 0.4 (*filled circle*), and 0.5 (*filled square*). Al-4.5%Cu-1.5%Mg (from [12])

of the apparent viscosity as a function of shearing time at a constant shear rate of $200\,s^{-1}$ and at various temperatures corresponding to various solid fractions. Steady state viscosity is reached after 1,000–2,000 s depending on the solid fraction.

Steady state viscosity was not observed in the case of the AZ91D magnesium alloy [13] sheared at $120\,s^{-1}$ at different temperatures corresponding to different solid fractions from 0.429 to 0.5. As the stirring time increases, the apparent viscosity initially decreases until it reaches a minimum value after which it starts to increase. This increase was attributed to the growth of the dendritic particles into spherical shapes.

In steady state, the rheological behavior has been found to be strongly shear thinning, the apparent viscosity decreasing with increasing shear rate (Fig. 6.5). Such a behavior has been confirmed on other semisolid systems and for higher shear rates. However, a tendency to observe saturation in the decrease of the viscosity with the highest shear rates has been reported [14].

The explanation for the shear thinning behavior of semisolid alloys is the decrease of the agglomerate size of the solid particles when the shear rate increases by rupture of the solid bridges. This decrease of the size leads to the release of the entrapped liquid, thus reducing the viscosity. Such an explanation has been confirmed at least partially by the work of Ito et al. [15] carried out on Al–Si alloy sheared at various rates. These authors confirmed that the concept of agglomerated particles is entirely justified through metallographic observations. In addition, they confirmed that high shear rates lead to a smaller volume fraction of entrapped liquid. More recently, observations carried out by X-ray tomography have confirmed that semisolid structures are highly agglomerated as soon as the solid fraction exceeds 0.3 or 0.4 [16]. Indeed, solid bridges easily form between slightly misoriented particles producing

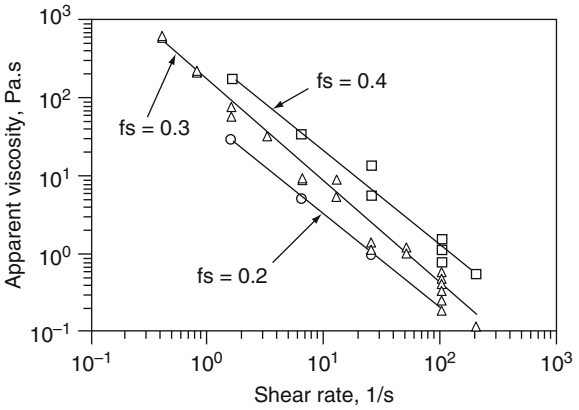

Fig. 6.5 Apparent viscosity of an A356 alloy as a function of the shear rate for various solid fractions (from [3])

a low angle grain boundary, which is not wetted by the liquid. The increase of the shear rate can therefore easily break the bridges so that the steady viscosity is the result of a dynamic equilibrium between the formation of bridges by contact of particles and diffusion, and their rupture induced by the shearing process.

The analysis of the slope of the curves reported in Fig. 6.5 shows that it is smaller than -1 (close to -1.3), which means that the shear stress decreases with increasing shear rate. Similar results were observed by other authors on Sn–Pb alloys [17, 18]. In their experiments, the steady state shear stress values were also measured by step changes in shear rate with sufficient time at each rate to reach a steady value of the torque. For McLelland et al., this anomalous behavior is interpreted in terms of rapid structural breakdown of particle agglomerates on increasing the shear rate from low initial values. For Koke and Modigell, this behavior can be explained at least partly by the change of the size of the globules. Indeed, when step changes in shear rate are carried out, the particle size is larger at the end of the experiment compared to the beginning and this explains at least partly the decrease in shear stress. These experiments therefore suggest that the rheological behavior at steady state depends on the way the material is tested.

Very recently, Kirkwood and Ward [19] demonstrated that the slope of the curve, such as that shown in Fig. 6.5 should be equal to $-4/3$. This value is proposed in conformity with the principle of shear reversal and it is independent of alloy system and fraction solid.

The viscosity at steady state has been related to the effective solid fraction [15], the effective solid fraction taking into account the liquid fraction entrapped in the agglomerates. Figure 6.6 shows that this viscosity depends only on this effective solid fraction without any influence of the shear rate. This result is not contrary to the idea of shear thinning since a similar value of the steady state viscosity can be obtained at two different shear rates corresponding to different volume fractions of entrapped liquid. The analysis of the straight line obtained for effective solid

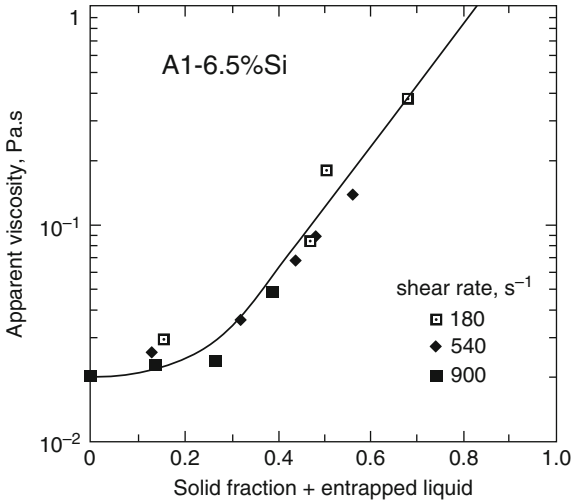

Fig. 6.6 Variation of the apparent viscosity at steady state as a function of the volume fraction of solid increased by the liquid volume fraction entrapped in the agglomerates for various shear rates (from [15])

fractions between 0.2 and 0.7 leads to a B value of 6.3, which is much smaller than that, about 10, obtained when the real solid fraction is considered (Table 6.1).

Another important aspect of the work of Joly and Mehrabian [2] is the study of the thixotropic behavior of semisolid Sn–Pb alloys. Thixotropy is defined as the time dependency of the viscosity. It reveals itself by the occurrence of hysteresis loops in the shear stress–shear rate diagram, when the shear rate is decreased until zero and increased again to its initial value after a given resting time. Figure 6.7 from the work of Joly and Mehrabian [2] shows such curves after various resting times. It is interesting to note that all the curves come back to the same initial point showing thus the reversible character of the microstructure of semisolid alloys. This result is obtained because the duration of the experiments is very short so that coarsening of the globules is not significant. In addition, coarsening of the globules is not a very important factor influencing the shear stress compared with the degree of agglomeration, provided that the size of the globules remains much smaller than the size of the specimen which is tested.

Other authors have used a similar procedure in the case of aluminum alloys containing SiC particles [20]. However, this procedure is not sufficient to fully characterize the agglomeration and deagglomeration kinetics, which are at the origin of the thixotropic behavior of semisolid slurries.

The transient evolution of stress observed after a step change of strain rate is a means to characterize these kinetics, provided that the tests are performed in very well controlled conditions (no slip at the walls during the shear rate change, rapid acquisition of the data, no inertial effects). Agglomeration processes are then dominant during step changes with decreasing shear rate values, whereas

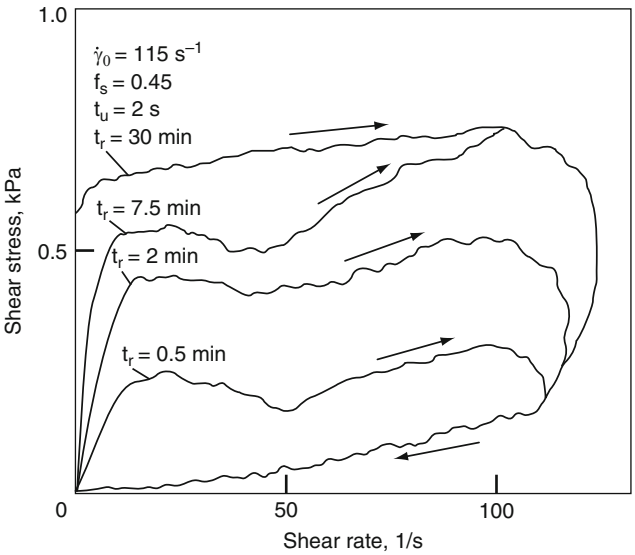

Fig. 6.7 Effect of the resting time t_r on the hysteresis loops of a Sn-15%Pb alloy; t_u is the time required to increase the shear rate up to its maximum value (from [2])

deagglomeration becomes dominating with increasing shear rate. Tests conducted in this way have shown that the alloy can exhibit a shear thickening behavior, viscosity increasing with increasing shear rate [21]. This shear thickening behavior was confirmed by Koke and Modigell [18], who studied the viscosity of a Sn-15.8%Pb alloy by using a Searle-type rheometer. By excluding inertial effects from the data evaluation, they obtained an isostructural flow curve with a shear stress depending on the shear rate to the power 2.07. The viscosity therefore increases almost linearly with increasing shear rate. Gautham and Kapur [22] observed also a shear thickening behavior under isostructural conditions, but the power law exponent was smaller, equal to 1.2.

The effect of the rest time on the shear stress measured during shear rates jumps has been determined by various authors [22, 23]. During the experiments, the shear rate was increased from 0 to a value typical of that experienced by the alloy when it enters the die cavity during an industrial thixoforming process. Increasing rest times lead to an increase in agglomeration and particle sizes thus increasing the shear stress recorded when the material is subjected to a shear rate jump from rest (Fig. 6.8). In addition, it was observed that the transient behavior of the slurry after a rapid change in shear rate occurs within 1 s of the shear-rate jump [23].

The increase of the resting time does not always lead to an increase of viscosity at a given shear rate. In the case of the AZ91D alloy, an increase of the viscosity was observed at high solid fraction (0.4), whereas at lower solid fraction, viscosity decreases slightly with increasing resting time [13]. The authors explained this result by different coarsening mechanisms. At high solid fraction, particle

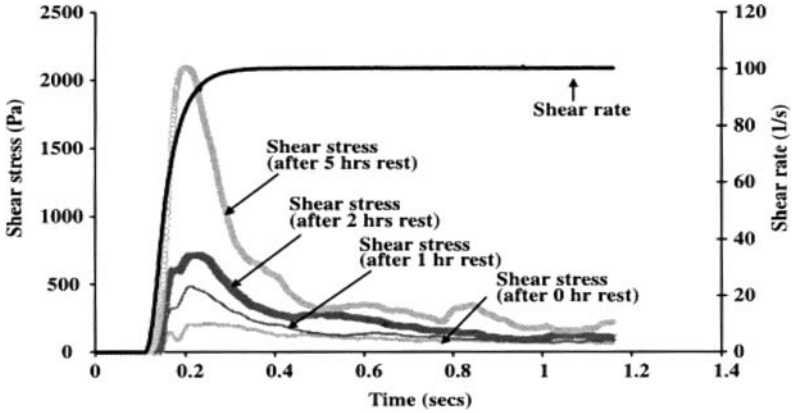

Fig. 6.8 Shear rate jumps from 0 to 100 s^{-1} after different rest times for Sn-15%Pb alloy at fraction solid 0.36 [23]

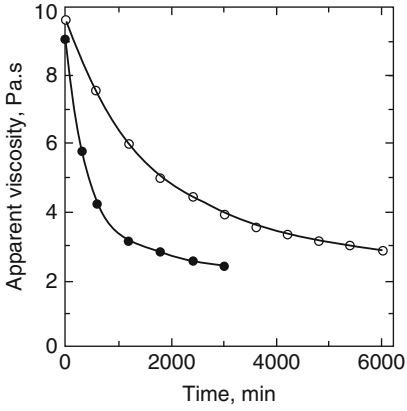

Fig. 6.9 Apparent viscosity as a function of the isothermal shearing time of an A356 alloy partially solidified ($f_s = 0.35$), (*open circle*) without and (*filled circle*) with 15% SiC particles added at time $t = 0$. The shear rate is 108 s^{-1} (from [11])

coalescence occurs preferentially to form larger ones in which the liquid entrapped in the individual particles remains entrapped. When the solid fraction is lower, the solid particles coarsen by Ostwald ripening and release the entrapped liquid into the bulk liquid. The effective liquid fraction is thus larger, which explains the viscosity decrease.

The results presented previously are concerned with semisolid alloys. Studies have been also carried out on alloys containing ceramic particles leading to similar results. However, it has been demonstrated that for particle fractions that are not too high, the viscosity of the alloy containing the ceramic particles can be lower than that of the alloy alone at the same temperature despite the presence of the particles (Fig. 6.9) [3, 11, 24]. This lower viscosity at higher solid fraction can be explained by the lower agglomeration level of the solid globules owing to the presence of the ceramic particles.

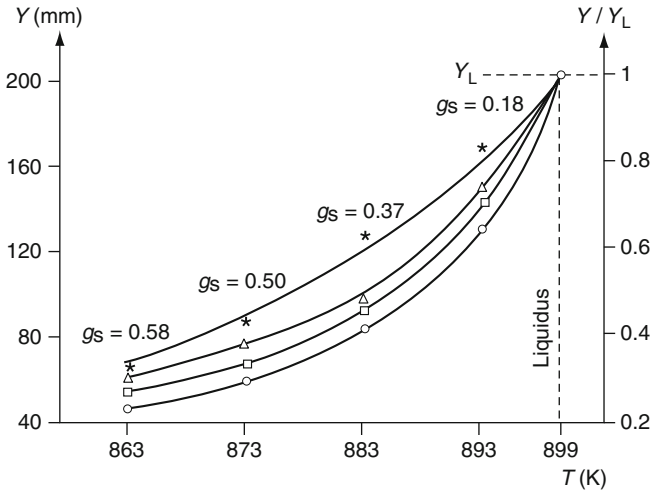

Fig. 6.10 Fluidity Y of an Al-10%Cu alloy in the semisolid state characterized by a solid fraction g_s. Y_L represents the fluidity of the liquid. The various curves are concerned with various stirring rates in revolutions/min. *open circle*: 340; *open square*: 482; *open triangle*: 695; *star sqaure*: 992 (from [25])

6.1.3 Results on Fluidity

As previously mentioned, the fluidity of an alloy is a technological parameter used by the foundry-man, who is pouring the metal in a mould to produce a part. Fluidity tests have been carried out with a partially solidified alloy subjected to stirring in comparison with the same alloy in the liquid state. The tests have shown that the alloy containing 50% solid still exhibits a relatively high fluidity and it is higher when the stirring rate before the test is increased [25] (Fig. 6.10). This result is in agreement with the shear thinning behavior of the alloy, the fluidity increasing (viscosity decreasing) with decreasing shear rate.

6.2 Partially Remelted Alloys

6.2.1 Experimental Methods

Shear Viscometer

During the shape forming of a billet by thixocasting, it experiences a very intense shear at the gate so that the capillary viscometer can be very interesting to obtain data about the rheological behavior of the billet in conditions close to forming. The capillary viscometer is similar to an extrusion apparatus in which the die is very

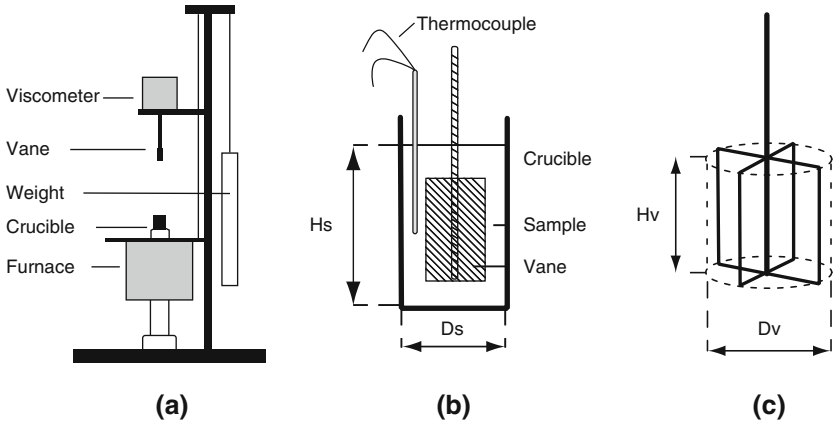

Fig. 6.11 Schematic drawing of the vane viscometer; (**a**) complete viscometer; (**b**) crucible with thermocouple and vane; (**c**) four bladed vane

wide. The pressure required to make the alloy flow in the tube at a given velocity is recorded and pressure sensors located along the tube allow the measurement of the pressure variations along the tube linked to the fluid flow. Such a device has been used by Paradies et al. [26] and Bernhard et al. [27].

The vane viscometer has been developed at NTNU Trondheim, Norway [28]. In a crucible placed in a furnace, the studied alloy is subjected to shear through the rotation of a four bladed vane (Fig. 6.11). This device can be used during solidification starting with the liquid alloy but it has been used mainly during partial remelting starting with the fully solid alloy. In this case, the specimen must be machined in order to allow the blades to be inserted in the alloy. The test consists of either imposing a shear at a given velocity, or imposing a stress during heating and recording the temperature (i.e., the liquid fraction) at which the material starts to deform.

Parallel Plate Compression

Parallel plate compression tests have been widely used to characterize the rheological behavior of semisolid alloys owing to the simplicity of the test. However, because friction cannot completely be avoided between the sample and the plates, the stress state is not purely uniaxial so that liquid segregation phenomena occur.

Two variants of the test have been used: the diameter of the specimen is smaller than that of the plates so that the current cross section of the specimen increases during the test or the specimen diameter is equal to that of the plates and in this case, the cross section remains constant which simplifies the analysis.

The problem of the flow of a fluid between two parallel plates has been solved many years ago. The solution is presented in [29] for the example of a Newtonian fluid, assuming sticky contact between the fluid and the plates. The Stefan equation is then obtained:

$$F = \frac{3\pi\eta R^4}{2h^3}\frac{\mathrm{d}h}{\mathrm{d}t}, \tag{6.1}$$

where F is the compression force, h is the instantaneous height of the specimen, R its radius, η is the viscosity of the fluid and $\frac{\mathrm{d}h}{\mathrm{d}t}$ is the compression rate.

More recently, a novel compression-type viscometer was developed [30–32]. The Drop forge viscometer has geometry similar to the parallel-plate viscometer, except the upper plate is suspended and then permitted to fall under the influence of gravity. As a result, the upper plate impacts the specimen at high velocity, thus leading to high shear rates of up to $10^4\ \mathrm{s}^{-1}$, which are representative of forming operations.

Direct Extrusion

The direct extrusion test consists of forcing a specimen of a given cross section to go through a die of smaller cross section under the action of a piston driven at a given velocity. This test is similar to the capillary viscometer and it reproduces quite well the conditions prevailing during thixocasting. The main drawback of the test is the friction between the piston and the container, which does not allow a precise determination of the rheology of the alloy. Nevertheless, experimental conditions leading to homogeneous deformation or liquid segregation can be defined by this test.

Back Extrusion

The previously mentioned drawback concerning friction is avoided if the test is performed in an inverse manner. The piston then plunges into the specimen, which flows between the container and the piston. This test is also close to industrial forming conditions and it allows the definition of the conditions for liquid segregation [33, 34]. Hence, the rheological behavior of the alloy in terms of apparent viscosity can be found, provided that some assumptions are made about the material and the boundary conditions [11].

Indentation

The indentation test used for characterizing semisolid alloys consists in the penetration of a tip of a given geometry into the specimen. It has been used at the beginning of the work on thixocasting to evaluate the softness of the material before starting the injection. It is therefore very interesting from this viewpoint, but it cannot easily lead to the rheology of the alloy in view of the very complex stress states that are generated under and in the vicinity of the tip. The analysis can be made by an inverse method using a computer code for the numerical simulation of the test [35, 36].

Drained Compression

In order to evaluate the compressibility of the solid, drained oedometric compression tests and drained triaxial compression tests have been carried out [37]. The drained oedometric compression consists of compressing the specimen in a container against a filter, which is very permeable for the liquid (Fig. 6.12). The strain state is well defined but this is not the case for the stress state, which can lead to some problems during the analysis of the test. The test allows also the measurement of the liquid volume fraction, which is really entrapped inside the solid. In addition, after complete filtration, a specimen is obtained, which has the composition of the solid phase in the semisolid alloy. The test can be performed without any major problem with industrial alloys, i.e., with alloys of relatively high solidus temperature.

The poor knowledge of the stress state in the previous test does not hold in the drained triaxial compression test, since the stress state is imposed and the volumetric strain of the sample is directly determined through the liquid quantity, which flows across the filter (Fig. 6.13). However, the specimen must be wrapped into a deforming envelope, which leads to problems in the case of industrial alloys. The

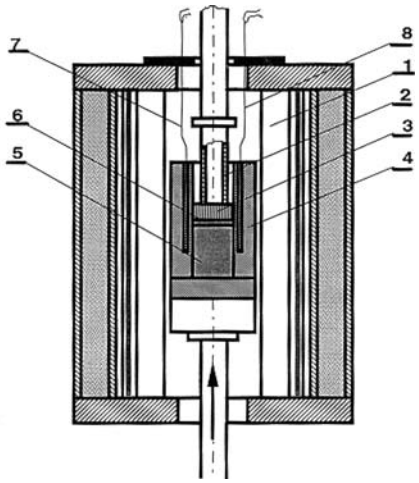

Fig. 6.12 Schematic drawing of the drained oedometric compression apparatus used for Al alloys. From [37]. 1: I.R. lamp furnace; 2: piston; 3: filter support; 4: container; 5: specimen; 6: fabric of SiC Nicalon fibers; 7, 8: thermocouples

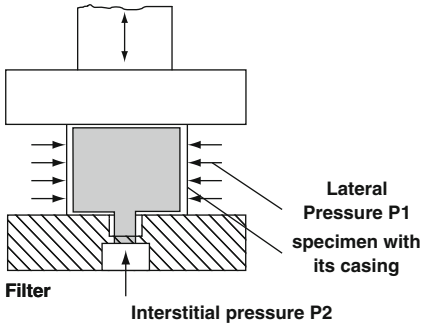

Fig. 6.13 Schematic drawing of the drained triaxial compression apparatus. From [37]

test has been applied successfully on low melting temperature alloys, such as Sn–Pb, the envelope being constituted of silicone rubber [37, 38]. It has also been used with the aluminum alloy A356. In this case, the envelope was made with pure Al and the pressure was applied by inert gas. The contribution of the envelope is then not negligible, which requires corrections of the applied axial stress [37].

Tension

The tensile test is normally not used in the case of semisolid alloys with a globular structure owing to the very low cohesion of the material when the solid fraction is typically that of billets suitable for thixocasting. However, in order to predict the liquid segregation phenomena, which can occur during a test, it is necessary to know the rheological behavior of the alloy for any stress states and higher solid fractions than those under normal conditions. Tensile tests have been carried out with magnesium alloys in particular [39].

6.2.2 Results in Terms of Apparent Viscosity

Influence of the Morphology of the Solid Phase

The mechanical tests carried out during partial remelting allow the influence of the morphology of the solid phase on the rheological behavior of the alloy to be easily determined. The tests are carried out, either after a predetermined holding time in the semisolid state for materials produced by various methods or subjected to various thermomechanical treatments, or after various holding times. These tests therefore allow various materials to be compared or they permit the determination of the optimum holding time in the semisolid state before the forming operation. Figure 6.14 shows the influence of the holding time on the shear stress deduced from

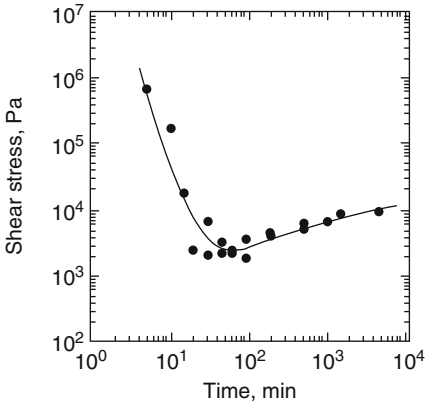

Fig. 6.14 Variation of the shear stress deduced from a compression test as a function of the isothermal holding time in the semisolid state before compression at 580°C of an A357 alloy partially remelted ($f_s = 0.45$). The compression rate is 0.01 s^{-1}. From [11]

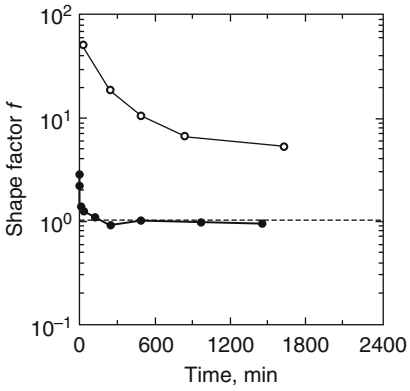

Fig. 6.15 Variation of the shape factor of the globule as a function of the isothermal holding time in the semisolid state at 580°C ($f_s = 0.45$) of an A357 alloy continuously cast (*open circle*) without and (*filled circle*) with electromagnetic stirring. The *dashed line* corresponds to perfect spheres. From [11]

a compression experiment carried out on an A357 aluminum alloy solidified initially with electromagnetic stirring. The shear stress (or viscosity) decreases sharply at short holding times to a minimum and then increases slightly when the holding time increases too much. The decrease of the stress is correlated with the globularization of the solid phase as shown by the concomitant decrease of the shape factor of the globules with increasing holding time (Fig. 6.15). This shape factor F is defined as:

$$F = \frac{1}{6\pi f_s} \frac{S_V^2}{N_A}, \tag{6.2}$$

where S_V is the globule–liquid interface surface area per unit volume and N_A represents the number of globules per unit area of section of the specimen. For long holding times, the increase of the viscosity is due to the agglomeration and growth of the globules, which become predominant over the globularization.

The entrapped liquid inside the globules plays a very important role in determining the viscosity of the alloy. This liquid does not participate in the rearrangement of the globules during deformation [15]. The alloy is thus equivalent to a material with a smaller liquid fraction, which therefore increases the viscosity. This increase is generally quite important owing to the very large sensitivity of the viscosity to the liquid fraction.

The degree of connectivity of the solid globules is also an important factor to take into account to describe the behavior of semisolid alloys. This connectivity can be quantified by the contiguity of the solid phase C_s or the contiguity volume $C_s f_s$ [40]. C_s is defined as $2N_L^{ss}/\left(2N_L^{ss} + N_L^{sl}\right)$ where $2N_L^{ss}$ is the number of interfaces between two solid globules per unit length and N_L^{sl} is the number of interfaces between the solid and the liquid per unit length (see the Microstructure Sect. 2.3, for a more complete discussion concerning contiguity). In the case of the electromagnetically stirred AA6082 tested by means of backward extrusion, these authors have shown that homogeneous deformation occurs when the contiguity volume is smaller than 0.3, whereas for larger values, the alloy loses its favorable flow characteristics. The influence of Ba addition was also studied and it was shown that Ba reduces the

solid–liquid interface energy leading to an increased penetration of the liquid phase between globules.

Influence of Shear Rate

Shear thinning steady state behavior

As for the alloys tested during solidification that exhibit a shear thinning behavior, the partially remelted alloys show a viscosity, which decreases as the shear rate increases. Such an evolution can be observed over a very wide range of shear rates as shown in Fig. 6.16 obtained with aluminum alloys and a magnesium alloy by compression tests at low shear rates and back extrusion tests at high shear rates [39]. The slope of the curve is close to –1 for the aluminum alloys, which means that the shear stress is almost independent of the shear rate. The slope is a little higher for the magnesium alloy. These curves are characteristic of a steady behavior, which results for the stirred alloys from equilibrium between agglomeration and deagglomeration of the globules.

Transient behavior

In order to determine the behavior during transient situations, step changes of strain rates can be performed during simple compression experiments. Figure 6.17 shows

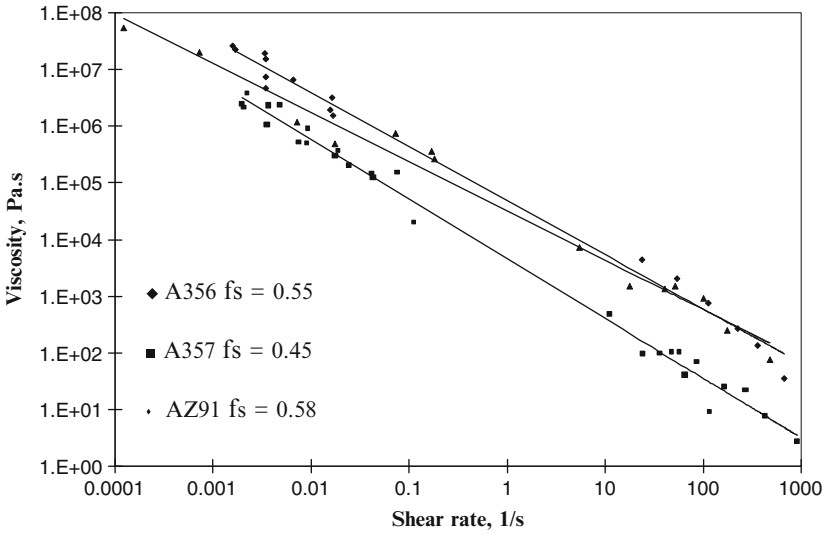

Fig. 6.16 Viscosity as a function of the shear rate for two aluminum alloys (A356, A357) and a magnesium alloy (AZ91) showing the shear thinning behavior of the alloys at solid fractions f_s close to 0.50. From [39]

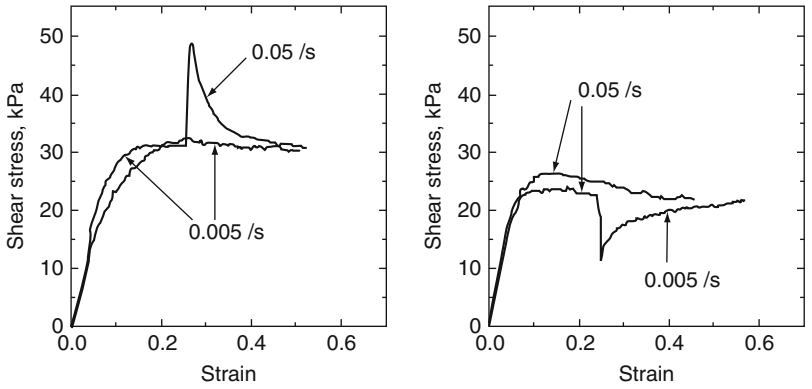

Fig. 6.17 Step changes of strain rate with increasing values (*left*) and decreasing values (*right*) for a Al-6%Si-0.6%Mg alloy deformed in compression at 580°C leading to a solid fraction of 0.55. From [11]

typical stress–strain curves resulting from step changes of strain rates with increasing and decreasing values. During an increase of strain rate, the stress increases rapidly and then decreases to a value close to that without strain rate change. Similarly, for a decrease of strain rate, the stress decreases sharply and then increases to the value close to that without step change. Such a behavior is in agreement with the previous result, which indicates that stress is almost independent of strain rate at steady state in this particular case. Conversely, the instantaneous change of the stress is characteristic of the behavior of the alloy at constant structure, i.e., at the degree of agglomeration of the globules, which is fixed by the strain rate applied before the strain rate change. The stress variation that follows immediately suppressed the step change is due to the deagglomeration of the globules for an increasing change and to the agglomeration for a decreasing change. As shown in Fig. 6.17, the strain for breakdown is smaller than that for buildup which is expected since breaking up of bonds between spheroidal solid particles in agglomerates is likely to be easier than the formation of bonds during shear rate drops.

The explanation in terms of microstructural changes is confirmed by tests with step changes of strain rate carried out on nonglobular structures on the one hand, and on aluminum matrix composites on the other hand. In the case of nonglobular structures, the stress increases during the strain rate change and shows a plateau characteristic of the new strain rate. The strain rate sensitivity of the stress is then characteristic of high temperature deformation of a solid.

A very similar behavior has been observed in the case of aluminum matrix composites containing SiC particles [42]. In these materials, the globular structure of the solid phase is obtained without any particular treatment owing to the presence of the SiC particles. Indeed, the particles are preferentially located in the liquid when the alloy is in the semisolid state, which allows a completely deagglomerated state of the structure (Fig. 6.18 [41]). It is to be noted, however, that this particular position of the particles is only possible for solid fractions, which are not too high

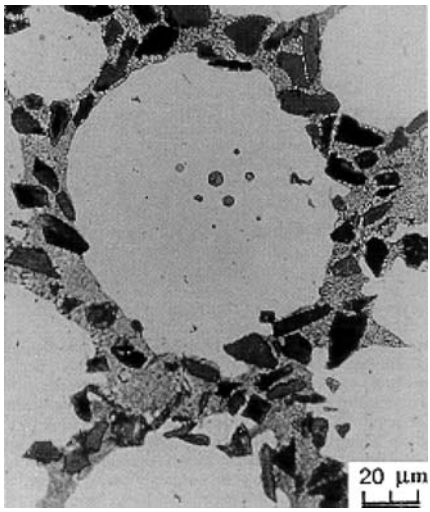

Fig. 6.18 Micrograph of a A356 + 20%SiC composite after partial remelting and quenching showing the position of the SiC particles in the eutectic mixture (liquid in the semisolid state). From [41]

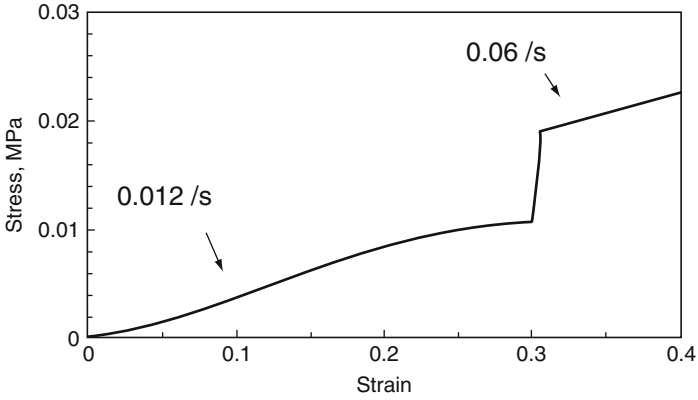

Fig. 6.19 Effect of a step change of strain rate in the case of a A356 + 20% SiC composite deformed in compression in the semisolid state. From [42]

and for a sufficient liquid fraction. During a step change of strain rate, a similar behavior to that with the nonglobular structure is observed (Fig. 6.19), because the degree of agglomeration of the globules does not evolve after the change.

Another important aspect related to the effect of shear rate is liquid segregation. This effect has been particularly observed in the case of simple compression experiments for which a free surface is present at the periphery of the specimen [32, 43]. At low compression rate, liquid segregation occurs quite extensively, whereas at very high rate, no segregation is observed. The explanation for this lack of segregation is that liquid flow requires very high pressures to occur so that homogeneous deformation is more likely to operate.

Table 6.2 Values of the parameter B in the expression $\eta = A\exp(Bf_s)$ for various alloys with globular structures

Alloy	Structure	Value of B	References
Al–Si	Globular	18–19	[11]
AZ91	Globular	15	[39]
Al–Ge	Globular	15	[44]
Sn-15%Pb	Globular	20.6	[45]

Influence of the Solid Fraction

The influence of the solid fraction on the viscosity of semisolid alloys has been determined by many authors on various alloy categories. Viscosity obviously increases with increasing solid fraction. As for the rheological behavior of the alloys during solidification, exponential laws have been proposed to account for this variation ($\eta = A\exp(Bf_s)$). Table 6.2 gives the various B values obtained with globular structures.

As for the alloys during solidification, the value of B is very high, which is consistent with the agglomeration of the globules leading to a larger effective solid fraction than the actual one.

The influence of the solid fraction on the viscosity has also been determined in the case of aluminum matrix composites. Similar to the behavior during solidification, the addition of ceramic particles reduces the viscosity of the semisolid alloy at the same temperature. This result can be explained by the fact that the ceramic particles hinder agglomerate formation between the particles [41].

6.2.3 Results in Terms of Constitutive Behavior

Many tests carried out on partially remelted alloys impose a stress state with an important hydrostatic component. During deformation of a specimen with a free surface, the liquid is subjected to a pressure gradient, which can lead to liquid segregation. This phenomenon occurs frequently during the compression test (particularly in the case of an important friction between the specimen and the compression plates) and during direct and backward extrusion. It can happen also during a forming operation of a component. In order to predict this phenomenon, it is necessary to develop models that account for the two-phase nature of the material and to identify these models by using tests involving a triaxial stress state. Oedometric compression and drained triaxial compression have been developed with this objective. In addition, even if liquid segregation occurs mainly under compressive stress states, it is still possible for local tensile stresses to develop inside a part during forming leading to liquid sucking.

Drained compression tests allow the determination of the densification behavior of the solid phase assuming that the pressure required to expel the liquid, is negligible. Figure 6.20 shows the variation of the filtration pressure as a function

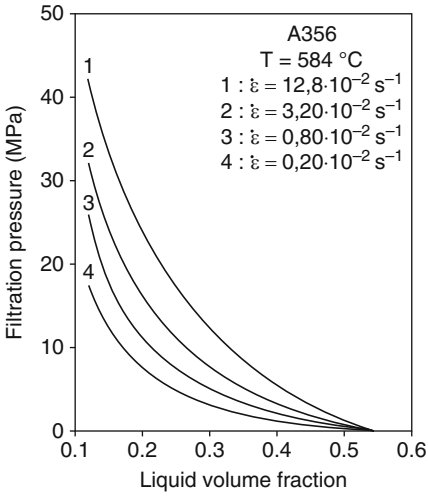

Fig. 6.20 Variation of the filtration pressure as a function of the liquid fraction remaining in the specimen during drained compression at various strain rates (from [46])

of the liquid fraction remaining in the specimen for an A356 alloy compressed at 584°C at various strain rates [46]. The filtration pressure increases with decreasing liquid fraction and increasing strain rate. These tests have demonstrated that a dendritic structure is more sensitive to densification than a globular structure at low solid fractions but the inverse is observed at high solid fraction [38]. They have also demonstrated that the liquid fraction really entrapped inside the solid globule is usually small (about 2%). In-situ X-ray microtomography carried out more recently on Al–Cu alloys has confirmed that the liquid fraction really present in the semisolid state can be quite different from that measured after solidification owing to the fact that quenching is usually not fast enough to freeze the microstructure [47].

Tensile tests have been carried out with an AZ91 magnesium alloy at high solid fractions to ensure sufficient cohesion of the solid phase. These tests have demonstrated that the tensile strength is much smaller than the compressive strength in the case of globular structures, whereas they are not so different in the case of dendritic structures [48]. This characteristic must be taken into account in the constitutive law of the semisolid alloy.

6.3 Comparison Between Partially Solidified and Partially Remelted Alloys

To the best of our knowledge, there is no detailed evaluation of the rheological behavior of the same alloy obtained by partial solidification on one hand and by solidification and subsequent partial remelting on the other hand. This comparison is obviously quite difficult to make, since it will depend on the way the alloy is partially solidified and the way it is reheated up to the temperature in the semisolid

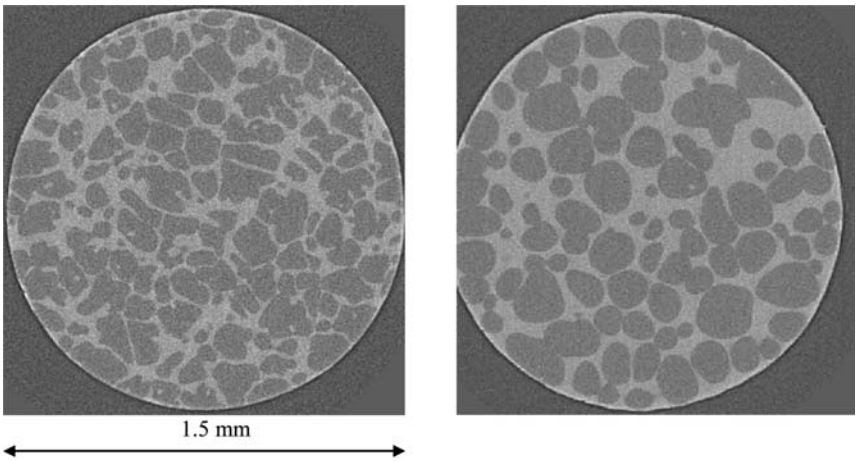

Fig. 6.21 Microstructures of an Al-15.8%Cu alloy after solidification (*left*) and after partial remelting at 555°C and holding for 80 min (*right*). These micrographs were obtained from in-situ tomography experiments carried out at ESRF Grenoble (from [50])

state for testing. In the case of conventional solidification, it has been observed that the liquid is more uniformly distributed in the interdendritic spaces than during remelting for which the liquid is more concentrated in the form of pockets. The strength of the alloy is thus smaller during partial solidification than during remelting at the same solid fraction [49]. In the case of globular structures, the situation is more complex, since strong evolution of the microstructure can take place during remelting. Solidification could lead directly to globules of the solid phase. These globules are well separated but they are not fully globular. Partial remelting leads to globularization of the microstructures but at the same time, agglomeration can occur. Figure 6.21 shows the microstructure of an Al-15.8%Cu alloy obtained during solidification with grain refinement and the same microstructure after partial remelting and holding for about 80 min in the semisolid range [50]. These microstructures were obtained by in-situ X-ray microtomography carried out at ESRF, Grenoble. It is clearly observed that the microstructure obtained during solidification is not fully globular, but most of the solid grains are isolated by a liquid film. After holding, the globules become more spherical (their shape factor decreases) but necks form in between the globules. The rheological behavior of these two types of microstructure is difficult to foresee: Globularization would decrease viscosity, whereas agglomeration would increase it.

6.4 Yield Stress

Several models used to describe the behavior of semisolid alloys include a yield stress, such as the Bingham or the Herschel–Bulkley equations (see Sect. 7.3). Basically, the yield stress is the stress below which there is no flow of the alloy.

Experiments to determine such a yield stress have been performed both on partially solidified and partially remelted alloys. Experiments were carried out by Modigell and Koke [51] by using a stress-controlled Couette-type rheometer. Increasing and decreasing linear stress ramps were used and they clearly observed no flow of the slurry below a given stress. This yield obviously depends on the material history. Other experiments have been performed by using a vane rheometer in which an alloy is progressively heated from the solid state [28]. The test consists in imposing a torque during heating and recording the temperature (i.e., the solid fraction) at which the material starts to deform. The shear stress is then calculated from this torque, which corresponds to the yield stress for this solid fraction.

Kirkwood and Ward [52] argue against the use of a yield point based on several experimental results at low shear rates [17, 23], which indicate that there is no limiting value of stress as shear rate decreases, viscosity and shear stress continuing to increase. However, the fact that there is no limiting value of stress when shear rate decreases is not similar to having a yield point. In the first type of experiments, a given nonzero stress must be applied to induce the flow of the slurry, whereas in the second, a shear rate is applied and it is observed that decreasing the shear rate leads to a continuous increase in shear stress due to the increased agglomeration of the globules. It is to be noted that this increase in shear stress with decreasing shear rate is observed if the slope of the viscosity vs. shear rate curve is smaller than -1, which is often the case in partially solidified alloys (Fig. 6.5). If this slope is greater than -1, stress will decrease with decreasing shear rate and in this case, extrapolation of the curve at zero shear rates could lead to a yield stress.

A serious objection against the existence of a yield stress is that deformation of a semisolid slurry takes place at very high homologous temperatures (T/T_L is close to 1 if T_L refers to the liquidus temperature of the alloy), so that diffusion will always take place without any yield stress. Therefore, a semisolid slurry will deform whatever the applied stress level but the resulting strain rate could be very small and even hardly detectable. This is obviously not interesting in practice so that the yield stress will be defined as the stress below which there is no detectable strain within the duration of the experiment. During injection, for which this duration is very small, the apparent yield stress could be quite large, whereas it could be much smaller during a laboratory experiment carried out at much smaller velocity. It seems therefore that the concept of yield stress is convenient to account for observations of regions described as unyielded or dead in a slurry having experienced a forming operation.

6.5 Concluding Remarks

Experimental results concerning the rheological behavior of semisolid alloys show that this behavior is very complex, since it involves many parameters which evolve during the experiment. These parameters are obviously linked both to the material (solid fraction, solid morphology, ...) and to the conditions of the experiments

(shear rate, deformation mode, ...). Although a huge number of experiments have been carried out so far on many types of materials, there is still debate concerning the type of experiments to be carried out, the interpretation of the results, and their modeling. In particular, for numerical simulation of forming processes, it is necessary to carry out experiments, which reproduce at best the forming conditions. Transient situations are therefore very important since the forming operations last usually less than a second. On the contrary, steady state conditions do not prevail during forming but they are nevertheless interesting from a more fundamental point of view.